

Photophoresis in a selectively excited gas

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A study is made of the drift of a macroscopic particle in a gas subjected to the field of monochromatic radiation. If the radiation interacts resonantly with the gas and if the interactions of excited and unexcited molecules with the surface of a particle are different, the particle experiences not only a radiometric, but also a light-induced force. The direction of the latter force is governed by the sign of the detuning of the radiation frequency from an atomic resonance. It is also shown that resonant excitation of molecules may alter considerably not only the magnitude, but also the direction of the radiometric force. Numerical estimates show that the light-induced component of the drift of a particle is several orders of magnitude stronger than the radiometric component of photophoresis.

A traveling light wave can induce a directional macroscopic flux of light-absorbing molecules present in a mixture with a buffer gas.^{1,2} The mechanism of appearance of such light-induced drift may be observed also in aerosols if they perform the role of a buffer gas.

We shall assume that a traveling light wave is absorbed as a result of an electronic or a vibrational-rotational transition from the ground (n) to an excited (m) state of a gas molecule and that the frequency of the light wave ω is close to the transition frequency ω_{mn} . In view of the Doppler effect, only those molecules interact with radiation which have velocities \mathbf{v} close to the resonance values, i.e., those which satisfy the condition $\mathbf{k}\cdot\mathbf{v} = \Omega \equiv \omega - \omega_{mn}$ (\mathbf{k} is the wave vector). The molecules that have absorbed radiation go over to the excited state. If the detuning of the radiation frequency from a resonance differs from zero ($\Omega \neq 0$), the distribution functions of the velocities of the excited (f_m) and unexcited (f_n) molecules are asymmetric relative to zero value of the Doppler factor ($\mathbf{k}\cdot\mathbf{v}$). Consequently, there are macroscopic, opposed, and collinear with the wave vector \mathbf{k} fluxes of excited \mathbf{J}_m and unexcited \mathbf{J}_n molecules. Since the gas as a whole is at rest, we have $\mathbf{J}_m + \mathbf{J}_n = 0$.

If the gas contains some macroscopic body and the excited and unexcited molecules interact with this body differently, then the opposed fluxes \mathbf{J}_m and \mathbf{J}_n flowing around the body experience different resistances. This creates an uncompensated force \mathbf{F}_s which is exerted by the gas on the body.

The force \mathbf{F}_s acting on a spherical macroscopic particle in a gas under free molecular flow conditions is calculated in Ref. 3 on the assumption of elastic specular-diffuse reflection of the molecules by the surface of the particle. It is assumed that the particle has an infinitely high thermal conductivity. We can therefore ignore the radiometric component F_R of photophoresis.⁴

The question of the influence of excitation, which is selective in respect of the molecular velocities, on the radiometric component of photophoresis requires a separate study. Moreover, it would be interesting to develop a theory with a generalized model of the boundary conditions.

We shall consider a spherical particle in a resonant gas. We shall assume that the radius R of this particle is much less than the mean free path of the molecules. Then, in the

zeroth approximation (in terms of the reciprocal of the Knudsen number) the kinetic equations for two-level systems² yield a relationship between distribution functions of excited and unexcited molecules:

$$f_m - \frac{\kappa}{2} (f_n - f_m) = 0, \quad (1)$$

where

$$\kappa = \frac{4|G|^2\Gamma}{\Gamma_m[\Gamma^2 + (\Omega - \mathbf{k}\mathbf{v})^2]}, \quad G = \frac{E_0 d_{mn}}{2\hbar};$$

κ is the saturation parameter; Γ is the homogeneous half-width of the absorption line; Γ_m is the radiative decay constant; d_{mn} is the matrix element of the dipole moment of the n - m resonance transition; E_0 is the amplitude of the electric field in a traveling optical wave.

If the origin of the spherical coordinate system (r, θ, φ) is placed at the center of the particle, then far from the particle the complete distribution function is

$$f_m + f_n = f_\infty (1 + 2c_z u_\infty), \quad (2)$$

where

$$f_\infty = n_\infty (m/2\pi k_B T_\infty)^{3/2} e^{-c^2}, \quad \mathbf{c} = (m/2k_B T_\infty)^{1/2} \mathbf{v}, \\ \mathbf{u}_\infty = (m/2k_B T_\infty)^{1/2} \mathbf{U}_\infty;$$

\mathbf{U}_∞ is the velocity of the incoming gas flux; n_∞ and T_∞ are the total density of the molecules and the temperature of the gas far from the particle; the z axis is directed along the wave vector \mathbf{k} .

It is assumed that the velocity of the arriving gas flux is well above the velocity of sound, so that Eq. (2) is presented in its linearized form. Moreover, we shall consider only the case of low values of the saturation parameter ($\kappa \ll 1$), which is justified for example in the case of a low radiation intensity. Then, in the approximation linear in κ , it follows from Eqs. (1) and (2) that the distribution functions of the excited and unexcited molecules far from the particle are

$$f_m = \frac{\kappa}{2} f_\infty, \quad f_n = f_\infty \left(1 + 2c_z u_\infty - \frac{\kappa}{2} \right). \quad (3)$$

Terms of the order of κu_∞ are omitted from the above expression.

The distribution functions of the molecules incident on a particle are not distorted in the Knudsen flow regime and

are described by Eq. (3). The distribution function of the reflected molecules is given by the following expressions⁵:

$$v_r f_i^+ = \int_{v_r' < 0} |v_r'| (P_{ii} f_i^- + P_{ji} f_j^-) dv', \quad v_r > 0, \quad i \neq j, \quad (i, j) = n, m, \quad (4)$$

where P_{ii} and P_{ji} are the scattering kernels for the case when there is no change in the state of the molecules and when a transition $j \rightarrow i$ takes place, respectively.

We shall assume that T_s is the unknown temperature of the particle surface, that α_{ji} is the probability of the $j \rightarrow i$ transition on collision of a molecule with the surface, and that consequently $(1 - \alpha_{ji})$ is the probability of an elastic collision ($j \rightarrow j$). Then, in the case of statistical independence of the $j \rightarrow i$ transition processes and a change in the molecular velocity $\mathbf{v}' \rightarrow \mathbf{v}$ due to a collision with the surface and subsequent diffuse scattering, we have⁵

$$P_{ii}^s(T_s) = (1 - \alpha_{ij}) P(T_s), \quad P_{ji}^s(T_s) = \alpha_{ji} P(T_s), \quad P(T) = \frac{v_r}{2\pi} \left(\frac{m}{k_B T} \right)^2 \exp\left(-\frac{mv^2}{2k_B T}\right), \quad (5)$$

whereas the distribution functions are described by

$$f_{is}^+ = (1 - \alpha_{ij}) f_i^s + \alpha_{ij} f_j^s, \quad (6)$$

$$f_i^s = n_i^s \left(\frac{m}{2\pi k_B T_s} \right)^{3/2} \exp\left(-\frac{mv^2}{2k_B T_s}\right),$$

$$i \neq j, \quad (i, j) = n, m.$$

The relationship between the temperature of the particle surface T_s and the corresponding partial densities n_i^s of the excited ($i = m$) and unexcited ($i = n$) molecules is determined by the condition that the whole of the gas is at rest:

$$\int_{v_r > 0} v_r (f_{ns}^+ + f_{ms}^+) dv = \int_{v_r < 0} |v_r| (f_n^- + f_m^-) dv. \quad (7)$$

In general, for an arbitrary nature of the gas-surface interaction the distribution functions of the reflected molecules can be represented conveniently in the form

$$f_i^+ = f_{is}^+ + \Delta f_i^+, \quad (8)$$

where

$$v_r \Delta f_i^+ = \int_{v_r' < 0} |v_r'| (\Delta P_{ii} f_i^- + \Delta P_{ji} f_j^-) dv', \quad v_r > 0, \quad \Delta P_{ii} = P_{ii} - P_{ii}^s, \quad \Delta P_{ji} = P_{ji} - P_{ji}^s.$$

It follows from the detailed-balance principle⁵ that for any scattering kernel we have

$$\int_{v_r' < 0} |v_r'| \Delta P_n(T_\infty) f_\infty dv' = 0, \quad \Delta P_i = \Delta P_{ii} + \Delta P_{ij}. \quad (9)$$

The temperature of the particle surface T_s is an unknown function of the polar angle θ . At low values of the radiation intensity or of the coefficient χ representing the absorption of light by the particle the relative difference between the temperatures of the particle and the gas is also small, i.e.,

$$\tau_s = (T_s - T_\infty) / T_\infty \ll 1.$$

Then, the scattering kernels, dependent on the particle sur-

face temperature, can be expanded as a Taylor series and we can retain only the linear approximation

$$\Delta P_i(T_s) = \Delta P_i(T_\infty) + \left(\frac{d\Delta P_i}{dT_s} \right)_{T_\infty} T_\infty \tau_s + \dots \quad (10)$$

The temperature field inside the particle is governed by the steady-state inhomogeneous heat conduction equation

$$-\lambda_p \Delta T_p(r, \theta) = Q_v(r, \theta), \quad (11)$$

where λ_p is the thermal conductivity of the particle and Q_v is the density of the internal sources of heat; in the case of planar monochromatic radiation, we have⁶

$$Q_v = 2n\chi kIB(r, \theta), \quad B(r, \theta) = |E(r, \theta)|^2 / E_0^2; \quad (12)$$

n is the refractive index, $E(r, \theta)$ is the local intensity of the electric field inside the particle, and I is the radiation intensity.

The methods of solution of the Mie problem and calculation of the function $B(r, \theta)$ are discussed in Ref. 7. The boundary conditions for Eq. (11) state that the temperature should be finite at the center of the particle $T_p(r=0) < \infty$ and that the radial heat fluxes at each point on the particle surface should be continuous

$$\left[\lambda_p \frac{\partial T_p}{\partial r} + \varepsilon \sigma (T_s^4 - T_\infty^4) + |q_r^+| - |q_r^-| \right]_{r=R} = 0, \quad (13)$$

where σ is the Stefan-Boltzmann constant; ε is the emissivity of the surface of the particle; the first term on the left-hand side of Eq. (13) represents the radial component of the heat flux vector inside the particle and the second represents thermal radiation; the radial components of the heat flux in the gas due to the molecules reflected or incident on the particle are described by the expressions

$$|q_r^\pm| = \frac{m}{2} \int_{v_r \leq 0} |V_r| V^2 (f_n^\pm + f_m^\pm) dv, \quad \mathbf{V} = \mathbf{v} - \mathbf{U}. \quad (14)$$

The force acting on the particle is found by direct calculation of the momentum transferred by the gas molecules to the particle by collisions

$$\mathbf{F} = \frac{\mathbf{k}}{|\mathbf{k}|} F, \quad F = m \int_{(S)} dS \left[\int_{v_r > 0} v_r v_z (f_n^+ + f_m^+) dv + \int_{v_r < 0} v_r v_z (f_n^- + f_m^-) dv \right]. \quad (15)$$

Here, S is the surface area of the spherical particle.

The general solution of Eq. (11) is⁴:

$$T_p(x, \theta) = \sum_{l=0}^{\infty} (a_l + b_l x) x^l P_l(\cos \theta), \quad x = r/R,$$

$$b_l = -\frac{R}{\lambda_p} \varphi_l, \quad (16)$$

$$\varphi_l = \frac{R}{2} (2l+1) \int_0^\pi \sin \theta P_l(\cos \theta) d\theta \int_0^1 x^{l+2} Q_v(x, \theta) dx,$$

where $P_l(\cos \theta)$ are Legendre polynomials. The unknown coefficients a_l are found by substituting Eq. (16) into the relationship (13) linearized with respect to τ_s and assuming

orthogonality between the expression obtained and the Legendre polynomials. Using Eqs. (3), (6)–(10), and (14), we obtain

$$\begin{aligned} \frac{1}{T_\infty}(a_i + b_i) = & \left\{ \frac{1}{2} \bar{v} p_\infty \left[1 + \pi^{1/2} \left(c^2, \left(\frac{d \Delta R_n}{dT_s} \right)_{T_\infty} \right) \right] \right. \\ & \left. + 4 \varepsilon \sigma T_\infty^4 \right\} A_i \delta_{0i} \\ & - \frac{\lambda_p}{R} b_i A_i - \frac{\pi^{1/2}}{2} \bar{v} p_\infty A_i \left\{ \frac{1}{4} u_\infty \delta_{1i} + 2 u_\infty \left[(c^2, c_r' \Delta R_n) \delta_{1i} \right. \right. \\ & \left. \left. - (c^2, c_\theta' \Delta R_n) (\sin \theta)^{(1)} \right] + \frac{1}{2} (c^2, \Delta R \kappa'^{(1)}) \right\}, \quad (17) \end{aligned}$$

where

$$\begin{aligned} A_i = & \left\{ \frac{1}{2} \bar{v} p_\infty \left[1 + \pi^{1/2} \left(c^2, \left(\frac{d \Delta R_n}{dT_s} \right)_{T_\infty} \right) \right] \right. \\ & \left. + 4 \varepsilon \sigma T_\infty^4 + \frac{\lambda_p}{R} T_\infty \right\}^{-1}, \\ (f, g) = & \frac{1}{\pi^{3/2}} \int_{c_r' > 0} f(\mathbf{c}) d\mathbf{c} \int_{c_r' < 0} |c_r'| \exp(-c'^2) g(\mathbf{c}, \mathbf{c}') d\mathbf{c}', \\ \kappa' = & \kappa(\mathbf{c}'), \\ f^{(1)} = & \frac{2l+1}{2} \int_0^\pi f P_l(\cos \theta) \sin \theta d\theta, \quad \bar{v} = \left(\frac{8k_B T_\infty}{\pi m} \right)^{1/2}, \\ \Delta R_i = & \left(\frac{2k_B T_\infty}{m} \right)^{1/2} \Delta P_i, \quad \Delta R = \Delta R_m - \Delta R_n; \end{aligned} \quad (18)$$

δ_{ik} is the Kronecker delta; $p_\infty = n_\infty k_B T_\infty$ is the equilibrium gas pressure.

In Eq. (17) we also allowed for the fact that the quantity ΔR_m should be attributed, in the linear (in respect of τ_s and κ) approximation, to the unperturbed gas temperature T_∞ and it should be regarded as independent of the angle θ .

The first term on the right-hand side of Eq. (17) governs the integral heating of the particle and the second the temperature inhomogeneity of its surface due to inhomogeneous distribution of the internal sources of heat as a result of absorption of electromagnetic radiation. In the third term the components proportional to u_∞ allow for the thermal polarization of the particle in an isothermal gas flow.⁸ Finally, the term containing κ represents the inhomogeneous heating of a particle by a light-induced isothermal heat flux. This effect is of interest for its own sake, because it is proportional to ΔR , consequently, it can be used to study the characteristic features of the interaction of the gas with the surface.

Now that we know the particle surface temperature, we can use Eqs. (3), (6)–(10), and (15) to find the force acting on the particle:

$$F = F_D + F_R + F_S, \quad (19)$$

where

$$F_D = -4\pi R^2 p_\infty \frac{u_\infty}{3} \left[\frac{\pi+8}{2\pi^{1/2}} - 4(c_r, c_r' \Delta R_n) - 8(c_\theta, c_\theta' \Delta R_n) \right] \quad (20)$$

is the restoring force,

$$\begin{aligned} F_R = 4\pi R^2 p_\infty \left[\frac{a_i + b_i}{12T_\infty} + \frac{2}{3} \frac{a_i + b_i}{T_\infty} \left(c_r, \left(\frac{d \Delta R_n}{dT_s} \right)_{T_\infty} \right) \right. \\ \left. - 2(\tau_s \sin \theta)^{(0)} \left(c_\theta, \left(\frac{d \Delta R_n}{dT_s} \right)_{T_\infty} \right) \right] \quad (21) \end{aligned}$$

is the radiometric force [$(a_i + b_i)$ follows from Eq. (16) if $l = 1$], and

$$F_S = 4\pi R^2 p_\infty \left[\frac{1}{3} (c_r, \Delta R \kappa'^{(1)}) - (c_\theta, \Delta R (\kappa' \sin \theta)^{(0)}) \right] \quad (22)$$

is the light-induced component of the photophoretic force.

The photophoretic velocity is found by equating the resultant force to zero. If we ignore the optical-pressure and gravity forces, we find that the condition in question can be written in the form

$$F_D + F_R + F_S = 0. \quad (23)$$

The difference between the interactions of the excited and unexcited molecules with the particle surface not only gives rise to the light-induced force of Eq. (22), but alters the radiometric component of Eq. (21).

By way of example, we shall consider a model of specular-diffuse reflection of molecules, according to which we have

$$\begin{aligned} \Delta P_{ii}(T) = (1 - \varepsilon_{ii})(1 - \alpha_{ij}) [-P(T) + \delta(\mathbf{v}' - \mathbf{v} + 2\mathbf{n}v_r)], \\ \Delta P_{ji}(T) = \alpha_{ji}(1 - \varepsilon_{ji}) [-P(T) + \delta(\mathbf{v}' - \mathbf{v} + 2\mathbf{n}v_r)], \end{aligned} \quad (24)$$

where \mathbf{n} is the normal to the surface; ε_{ij} and ε_{ji} are the fractions of the diffusely scattered molecules without a change in the state ($i \rightarrow j$) and after the transition ($j \rightarrow i$), respectively.

The restoring force is

$$\begin{aligned} F_D = -4\pi R^2 p_\infty u_\infty \frac{\pi^{1/2}}{6} \left(\frac{8}{\pi} + \varepsilon_n \right), \\ \varepsilon_i = \varepsilon_{ii} - \alpha_{ij} (\varepsilon_{ii} - \varepsilon_{ij}). \end{aligned} \quad (25)$$

Simple analytic expressions for the radiometric and light-induced forces can be obtained only in two cases.

1. *Homogeneous broadening* ($\Gamma \gg k\bar{v}$); this case is encountered near the long-wavelength edge of the infrared region. If $|\Omega| \gtrsim \Gamma$, we obtain

$$\begin{aligned} F_R = -4\pi R^2 p_\infty \varepsilon_n \frac{A_1}{12} \left[-\varphi_1 + \frac{\pi^{1/2}}{8} \bar{v} p_\infty (\varepsilon_n u_\infty + \pi^{1/2} \Delta \varepsilon \Omega \bar{\kappa}) \right], \\ F_S = 4\pi R^2 p_\infty \frac{\pi}{6} \Delta \varepsilon \Omega \bar{\kappa}, \quad \Delta \varepsilon = \varepsilon_n - \varepsilon_m, \quad \varphi_1 = J J_1, \\ \bar{\kappa} = \frac{|G|^2 \Gamma k \bar{v}}{\Gamma_m (\Gamma^2 + \Omega^2)^2}, \end{aligned} \quad (26)$$

$$A_1 = \left(\frac{1}{2} \bar{v} p_\infty \varepsilon_n + 4 \varepsilon \sigma T_\infty^4 + \frac{\lambda_p}{R} T_\infty \right)^{-1},$$

where J_1 is the asymmetry factor of the particle surface temperature.^{4,6}

The photophoretic particle velocity is found from Eqs. (23), (25), and (26):

$$U_{\phi} = U_S + U_R, \quad U_S = \frac{\pi}{2} \bar{v} \alpha \Delta \varepsilon \Omega \bar{\kappa},$$

$$U_R = \frac{\alpha}{4} \varepsilon_n A_1 \bar{v} \left(\varphi_1 - \frac{\pi}{8} \bar{v} p_{\infty} \Delta \varepsilon \Omega \bar{\kappa} \right), \quad (27)$$

$$\alpha = \left(\frac{8}{\pi} + \varepsilon_n + \frac{A_1}{16} \varepsilon_n^2 \bar{v} p_{\infty} \right)^{-1} \approx \left(\frac{8}{\pi} + \varepsilon_n \right)^{-1}.$$

2. *Inhomogeneous broadening* ($\Gamma \ll k\bar{v}$) is most typical of low-pressure gasses. If $|\Omega| \leq \Gamma$, we obtain

$$F_R = -4\pi R^2 p_{\infty} \varepsilon_n \frac{A_1}{12} \left[-\varphi_1 + \frac{\pi^{1/2}}{2} \bar{v} p_{\infty} \left(\frac{\varepsilon_n}{4} u_{\infty} + \frac{4}{\pi^{1/2}} \Delta \varepsilon \frac{\Omega}{\Gamma_m} \kappa_0 \right) \right],$$

$$F_S = 4\pi R^2 p_{\infty} \frac{4}{3} \Delta \varepsilon \frac{\Omega}{\Gamma_m} \kappa_0, \quad \kappa_0 = \left(\frac{|G|}{k\bar{v}} \right)^2. \quad (28)$$

The components of the photophoretic velocity of the particle are

$$U_S = 4\alpha \bar{v} \Delta \varepsilon \frac{\Omega}{\Gamma_m} \kappa_0, \quad (29)$$

$$U_R = \frac{\alpha}{4} \bar{v} \varepsilon_n A_1 \left(\varphi_1 - 2\bar{v} p_{\infty} \Delta \varepsilon \frac{\Omega}{\Gamma_m} \kappa_0 \right).$$

In any case the direction of the light-induced component of the force, calculated for a fixed value of $\Delta \varepsilon$, is governed only by the sign of the detuning Ω and is independent of the direction of propagation of radiation. If $\Delta \varepsilon > 0$, then for $\Omega > 0$ the direction of the velocity vector \mathbf{U}_S is identical with the direction of the wave vector \mathbf{k} , whereas for $\Omega < 0$ the vector \mathbf{U}_S is opposite to \mathbf{k} .

The magnitude and direction of the radiometric component of the photophoretic velocity \mathbf{U}_R depends on the temperature inhomogeneity of the particle, on the nature of the gas-surface interaction, and on the direction of the thermal polarization of the particle. All of them are governed by the parameters ε_m , ε_n , and the asymmetry factor J_1 , which may be negative, positive, or zero⁶ (for an absolutely absorbing particle we have $J_1 = -1/2$) and the sign of the detuning Ω .

We shall now give numerical estimates in order to compare U_S and U_R . Let us assume that $p_{\infty} \approx 1$ Torr, $T_{\infty} \approx 300$ K, $I \approx 1$ W/cm², $\lambda_p \approx 1$ W·m⁻¹·K⁻¹, $J_1 = -1/2$, $G \approx 10^8$ Hz, $\Omega \approx \Gamma \approx \Gamma_m \approx 10^7$ Hz, $\bar{v} \approx 10^3$ m/s, $\varepsilon_n \approx 0.9$, and $\varepsilon_m \approx 0.8$. Then in the homogeneous broadening case ($k \approx 10^3$ m⁻¹), we find that $U_R \approx 10^{-3}$ m/s and $U_S \approx 10^2$ m/s. In the inhomogeneous broadening case (when $k \approx 10^7$ m⁻¹), we obtain $U_R \approx 10^{-3}$ m/s and $U_S \approx 10^{-1}$ m/s. Therefore, the light-induced component of the photophoretic velocity exceeds by several orders of magnitude the radiometric component. It should also be mentioned that in the case of homogeneous broadening each term in Eq. (27) makes an approximately the same contribution to the value of U_R , whereas in the inhomogeneous broadening case we can ignore the second term in Eq. (29), since its contribution to U_R is only $\sim 1\%$.

Obviously, in the case of weakly absorbing particles ($J_1 \approx 0$) the main contribution to the radiometric components of the force to the photophoretic velocity comes from the terms which are due to the resonant interaction of a gas with light.

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