MOTION OF SMALL AEROSOL PARTICLE

IN A LIGHT FIELD

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A theory is derived for the photophoretic motion of a small aerosol particle, whose radius is much smaller than the wavelength of the light. The radiation pressure is taken into account; the effects of the incomplete accommodation of the energy and momentum of the gas molecules in collisions with the surface of the particle are also taken into account.

When illuminated, a spherical particle suspended in a gas experiences two forces: the radiation pressure and the photophoretic force, which arises because of the interaction of the molecules of the surrounding gas with the surface of the particle, which is heated nonuniformly by the light. The gas molecules reflected from the hot side of the particle after an inelastic collision move faster than those reflected from the cold side; as a result, the particle acquires a net momentum.

The motion of an aerosol particle in a light field was discovered by Ehrenhaft [1, 2]. Since the discovery, this effect has been studied in many experiments (see the review in [3]). The development of high-power lasers has stimulated further research in this field [4-7]. With laser radiation it is possible to achieve optical levitation of particles in air or vacuum, to accelerate electrically neutral particles to high velocities, and to sort particles according to size.

Debay [8] and Rubinowicz [9] have taken up the theory of the forces acting on a spherical particle in a light field. Rubinowicz [9] derived an equation for the photophoretic force acting on a spherical particle in an extremely nonrigorous manner, from the Knudsen equation for the force acting between two heated plates. The result is applicable only in the case of a small deviation of the surface temperature of the particle from the gas temperature at infinity, and this result also ignores the fact that some of the molecules undergo specular reflection at the surface. Furthermore, Rubinowicz [9] did not examine the velocity of the particle under the influence of the photophoretic force, so that a final theory for photophoresis could not be worked out.

Below we derive rigorous expressions for the photophoretic force in the free-molecular regime for an arbitrary surface temperature of the particle; we take into account the incomplete accommodation of the energy and momentum of the gas molecules in collisions with the surface. We also take into account the influence on the particle of the radiation pressure, and we derive the steady-state velocity.

1. Distribution of Absorbed Energy within a Sphere

Mie [10] has derived an exact solution for the problem of the distribution of the electromagnetic field within a homogeneous spherical particle and in the space near the particle:

$$E_r^{i,e} = E_0 \cos\varphi\,(k_{i,e}r)^{-2}\sum_{l=1}^{\infty} l\,(l+1)\,C_l^{i,e}P_l^{(1)}(\cos\theta_1)\,\varphi^{i,e}(k_{i,e}r),$$

$$E_\theta^{i,e} = -E_0\cos\varphi\,(k_{i,e}r)^{-1}\sum_{l=1}^{\infty}\left\{C_l^{i,e}P_l^{(1)\prime}\varphi_l^{i,e\prime}\sin\theta_1 - iB_l^{i,e}\varphi_l^{i,e}P_l^{(1)}\frac{1}{\sin\theta_1}\right\}, \quad (1)$$

$$E_\varphi^{i,e} = -E_0\sin\varphi\,(k_{i,e}r)^{-1}\sum_{l=1}^{\infty}\left\{C_l^{i,e}P_l^{(1)\prime}\varphi_l^{i,e\prime}\frac{1}{\sin\theta_1} - iB_l^{i,e}\varphi_l^{i,e}P_l^{(1)}\sin\theta_1\right\}.$$

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Here the superscript "i" corresponds to the internal fields, while "e" refers to the external fields. Here $k_i = k_0 N$, $k_e = k_0 = 2\pi/\lambda_0$, $N = n - i\varkappa$ is the complex refractive index, λ_0 is the light wavelength, $\varphi_l^i(z) \equiv \psi_l(z) = \sqrt{\frac{\pi z}{2}}\, J_{l+1/2}(z)$, $\varphi_l^e(z) \equiv \zeta_l(z) = \sqrt{\frac{\pi z}{2}}\, H_{l+1/2}(z)$, $J_{l+1/2}$ and $H_{l+1/2}$ are the Bessel and Hankel functions, $P_l^{(1)}$ is the associated Legendre polynomial, $\theta_1 = \pi - \theta$, and θ is the angle between the radius vector and the propagation direction of the light. The coefficients $C_l^{i,e}$ and $B_l^{i,e}$ are given by

$$
\begin{gathered}
C_l^i = iNc_l; \quad C_l^e = [\psi_l(\rho)\psi_l'(N\rho) - N\psi_l'(\rho)\psi_l(N\rho)]\, c_l, \\
B_l^i = iNb_l; \quad B_l^e = [\psi_l'(\rho)\psi_l(N\rho) - N\psi_l(\rho)\psi_l'(N\rho)]\, b_l, \\
c_l = [\zeta_l(\rho)\psi_l'(N\rho) - N\zeta_l'(\rho)\psi_l(N\rho)]^{-1}, \\
b_l = [\zeta_l'(\rho)\psi_l(N\rho) - N\zeta_l(\rho)\psi_l'(N\rho)]^{-1},
\end{gathered} \tag{2}
$$

where $\rho = k_0 R$. Below we will be concerned with only particles whose radii are much smaller than the wavelength of the incident radiation ($\rho \ll 1$), so that Eqs. (1) and (2) simplify. We are primarily interested in the two limiting cases of a weakly absorbing particle ($\varkappa\rho \ll 1$) and a strongly absorbing particle ($\varkappa\rho \gg 1$).

In the case of a weakly absorbing particle, the asymptotic expressions for the functions ζ_l and ψ_l for $\rho \ll 1$, $\varkappa\rho \ll 1$ are [10]

$$
\begin{gathered}
\zeta_l(\rho) = (2l-1)!!\, \frac{i-\rho}{\rho^l} \exp(-i\rho), \\
\psi_l(kr) = (kr)^{l+1}\,[2(2l+3) - k^2r^2]\, \frac{1}{2(2l+3)!!}\,.
\end{gathered} \tag{3}
$$

Substituting (3) into (1) and (2), we find $|E|^2$:

$$
|E(r,\theta)|^2 = 3E_0^2|3 + 2N^2|^{-2}\{3|3 + 2N^2|^2 + 2n\varkappa k_0 r\cos\theta_1(|3 + 2N^2|^2 - 5)\}. \tag{4}
$$

In deriving Eq. (3) we assumed the incident wave to be unpolarized, and we averaged the expression for $|E|^2$ over the angle φ. We see from (4) that the heat-source density, which is proportional to $|E|^2$, is distributed nearly uniformly over the volume of the particle ($\varkappa\rho \ll 1$). More heat is evolved at the illuminated side of the sphere ($\theta_1 < \pi/2$) than at the shaded side.

In the case of a strongly absorbing particle, the field penetrates a distance on the order of $(\varkappa k_0)^{-1} \ll R$ into the sphere, and the absorption is essentially superficial. In the case $\varkappa\rho \gg 1$, the asymptotic expression for ψ_l is [13]

$$
\psi_l(kr) = (-i)^l \exp(ikr)\left\{-0{,}5i + \frac{4(l+0{,}5)^2 - 1}{16kr}\right\}. \tag{5}
$$

Substituting (3) and (5) into (1) and (2), we find

$$
|E(r,\theta)|^2 = E_0^2 \exp[-2\varkappa k_0(R-r)]\, \frac{1}{2|N|^2}\left\{\frac{9}{2} + \frac{25}{36}(1 - 2\cos^2\theta_1)^2 + \frac{9n}{4|N|^2}\cos\theta_1\right\}. \tag{6}
$$

We see the exponential decay of the field into the interior of the particle typical of a strongly absorbing particle. It is interesting to compare (6) with the result obtained for a strongly absorbing particle of large size ($\rho \ll 1$). On the shaded side of a large sphere there is no light absorption ($|E|^2 = 0$), while for a small particle the values of $|E|^2$ on the illuminated and shaded sides are nearly the same ($n \ll \varkappa$). The absorption at the shaded side of the sphere occurs because of diffraction and is particularly important for small particles ($\rho \ll 1$).

2. Solution of the Heat-Conduction Equation

We consider the case in which the radius of the particle is much smaller than the mean free path of the gas molecules; in other words, we consider the free-molecular regime of the interaction of the gas with the surface of the particle. In this case the distribution function of the gas molecules incident on the particle is not distorted by those reflected from the particle:

$$
f^- = n_\infty\left(\frac{m}{2\pi kT_\infty}\right)^{3/2} \exp\left[-\frac{m(\mathbf{v} + \mathbf{u})^2}{2kT_\infty}\right], \tag{7}
$$

where n_∞ and T_∞ are the density of molecules and the gas temperature at infinity, and $\mathbf{u}$ is the velocity of the particle. Here the superscript " – " indicates that the projection of the velocity of the molecule onto the normal to the surface, v_n, is negative. We assume that a fraction q of the molecules are reflected from the surface in a diffuse manner, with an isotropic Maxwell distribution, while the rest $(1 - q)$ undergo specular reflection:

$$f^+ = qn_r \left(\frac{m}{2\pi kT_r}\right)^{3/2} \exp\left[-\frac{mv^2}{2kT_r}\right] + (1-q) f^- [\mathbf{v} - 2\mathbf{n}(\mathbf{nv})], \tag{8}$$

where $T_r = (1-\gamma)T_\infty + \gamma T_s$, and T_s is the surface temperature of the particle. The quantity q is usually called the "momentum-accommodation coefficient," while γ is the "energy-accommodation coefficient" [4]. The temperature distribution within the particle is determined from the solution of the steady-state heat-conduction equation

$$\varkappa_i \Delta T_i = \operatorname{div} \mathbf{I} = -2n\varkappa k_0 I B(r, \theta), \tag{9}$$

where $\varkappa_i$ is the thermal conductivity of the particle, $\mathbf{I} = \frac{c}{8\pi} \mathrm{Re}[\mathbf{E}, \mathbf{H}^*]$ is the Poynting vector, $B(\mathbf{r}, \theta) = |\mathbf{E}(\mathbf{r})|^2/E_0^2$, and $I = cE_0^2/8\pi$ is the energy flux density in the incident electromagnetic wave. The boundary conditions for Eq. (9) are the condition that the surface of the particle is impenetrable for the gas molecules and the continuity condition on the heat flux at the particle surface:

$$\sum_{\pm} \int_{\pm} d\mathbf{v}\,(\mathbf{nv}) f^{\pm}(\mathbf{v}, R, \theta) = 0, \tag{10}$$

$$-\varkappa_i \frac{\partial T_i}{\partial r}\Big|_{r=R} = \sum_{\pm} \int_{\pm} d\mathbf{v}\,(\mathbf{nv}) \frac{mv^2}{2} f^{\pm}(\mathbf{v}, R, \theta). \tag{11}$$

The superscripts "+" and "−" in the integral indicate that the integration is to be carried out over the half-spaces $v_n > 0$ and $v_n < 0$.

Solving Eq. (9) with boundary conditions (10) and (11), we find the following equation for the surface temperature of the particle:

$$T_s = \bar{T}_s + \sum_{l=1}^{\infty} \frac{J_l IRP_l(\cos\theta)}{l\varkappa_i + Rq\gamma n_\infty \sqrt{2kT_\infty/\pi m}}, \tag{12}$$

$$\bar{T}_s = T_\infty + \frac{Ik_a}{4kq\gamma n_\infty \sqrt{2kT_\infty/\pi m}}, \tag{13}$$

where

$$J_l = (2l+1)\, n\varkappa\rho \int_0^1 dx x^{l+2} \int_0^\pi d\theta \sin\theta P_l(\cos\theta) B(r, \theta),$$

$x = r/R$, $\bar{T}_s$ is the average surface temperature, and $k_a = 4J_0$ is the absorption factor, which is equal to the ratio of the energy absorbed per unit time to the energy flux incident on the geometric cross section of the sphere. For most substances the second term in the denominator in (12) is much smaller than the first and can be neglected.

3. Forces Exerted on a Particle in a Light Field. Velocity

The force exerted on the particle by the gas is equal to the resultant momentum transferred to the particle by the gas molecules which collide with the surface of the particle in a unit time:

$$\mathbf{F} = -\sum_{\pm} R^2 \int d\Omega_n \int_{\pm} d\mathbf{v}\,(\mathbf{nv})\, m\mathbf{v} f^{\pm}(\mathbf{v}, R, \theta) = -\pi R^2 p_\infty \left\{ q \int_{-1}^{+1} dy\, y \sqrt{\frac{T_r}{T_\infty}}\, \mathbf{e}_z + \frac{2m\left(8 + q\pi\sqrt{\bar{T}_r/T_\infty}\right)}{18\pi kT_\infty} \mathbf{u} \right\}, \tag{14}$$

where $y = \cos\theta$, $p_\infty = n_\infty kT_\infty$ is the gas pressure at infinity, and $\mathbf{e}_z$ is the unit vector along the light-propagation direction. In deriving Eq. (14) we made use of (7), (8), and (10), and we assumed $u \ll \sqrt{2kT_\infty/m}$. The first term is the photophoretic force exerted on the immobile particle, and the second term is the drag force. We see from (14) that if the temperature of the illuminated part of the sphere $(-1 < y < 0)$ is higher than that of the shaded part the photophoretic force is directed along the z axis (this is positive photophoresis). In the opposite case, the force is directed opposite the incident radiation (negative photophoresis). Substituting the solution of heat-conduction equation (12) into (14), we find the photophoretic force to be

$$\mathbf{F}_{\mathrm{p}} = -\pi R^2 p_\infty q \gamma I R J_1 \frac{\mathbf{e}_z}{3\varkappa_i \sqrt{\bar{T}_r T_\infty}}. \tag{15}$$

The radiation pressure is [8, 11]

$$\mathbf{F}_{\mathrm{rp}} = I k_{\mathrm{rp}} \pi R^2 \mathbf{e}_z, \tag{16}$$

where $k_{\mathrm{rp}} = k_a + k_c - q_d$ is the radiation-pressure factor, and k_a and k_c are the absorption and scattering factors. The quantity k_a is given by Eq. (13); k_c and q_d are given by the following equations [11]:

$$k_c = \sum_{l=1}^{\infty} \frac{2\, l^2 (l+1)^2}{\rho^2 (2l+1)} \left(|C_l^e|^2 + |B_l^e|^2\right), \tag{17}$$

$$q_d = -\frac{4}{\rho^2} \operatorname{Re} \sum_{l=1}^{\infty} \frac{l(l+1)^2}{2l+1} \left[C_l^{e*} B_l^e - i \frac{l(l+2)}{2l+3} \left(C_l^{e*} C_{l+1}^e - B_l^{e*} B_{l+1}^e \right) \right]. \tag{18}$$

Let us consider in more detail the cases of weakly and strongly absorbing particles.

I. Weakly Absorbing Particle. To derive an equation for the photophoretic force, we substitute Eqs. (12), (9), and (4) into (15):

$$\mathbf{F}_{\mathrm{p}} = \frac{\pi R^4 p_\infty \gamma q I k_a n \varkappa k_0 \mathbf{e}_z}{30 \varkappa_i \sqrt{\bar{T}_r T_\infty}} \left(1 - \frac{5}{3 + 2|N|^2}\right), \tag{19}$$

$$k_a = \frac{24 n \varkappa \rho}{|2 + N^2|^2}, \quad k_{\mathrm{rp}} = k_a. \tag{20}$$

These equations are found by substituting Eqs. (2) into (13), (16), (17), and (18) and using (3) and (4). With $q = 1$ and $\bar{T}_S = T_\infty$, Eq. (19) is the same as the equation derived for the photophoretic force in [9]. We see from Eq. (19) that the photophoretic force acting on a small, weakly absorbing particle always acts along the radiation-propagation direction (positive photophoresis). Setting the total force acting on the particle equal to zero, and using (16), (19), and (20), we find the steady-state velocity to be

$$\mathbf{u} = \sqrt{\frac{2\pi}{m k \bar{T}_r}} \frac{I k_a R^2 q \gamma n \varkappa k_0 (1 + \alpha)\, \mathbf{e}_z}{20 \varkappa_i \left(8 + q\pi \sqrt{\bar{T}_r / T_\infty}\right)} \left(1 - \frac{5}{3 + 2|N|^2}\right), \tag{21}$$

where

$$\alpha = \frac{F_{\mathrm{rp}}}{F_{\mathrm{p}}} = \frac{30 \varkappa_i \sqrt{\bar{T}_r T_\infty}\, (3 + 2|N|^2)}{R^2 c p_\infty \gamma q n \varkappa k_0 (3 + 2|N|^2 - 5)}.$$

If $\alpha \ll 1$, the velocity is proportional to the cube of the radius of the particle, so that light can be used to sort the particles of a polydisperse aerosol according to size. If $\alpha \gg 1$, the velocity is proportional to the first power of the radius, and the sorting is less efficient.

II. Strongly Absorbing Particle. Substituting Eqs. (2), (3), (5), and (6) into (13), (16)-(18), we find

$$k_a = \frac{6n}{|N|^2}; \quad k_{\mathrm{rp}} = \frac{6n}{|N|^2} + \frac{14}{3} \rho^4. \tag{22}$$

The condition for strong absorption ($\varkappa\rho \gg 1$) in the case of small particles ($\rho \ll 1$) is usually satisfied only for metals with a high electrical conductivity. We know that the heat in metals is transferred by free electrons, so that metals which are good conductors have a high thermal conductivity. The photophoretic force is inversely proportional to the thermal conductivity, and, as estimates show, this force is always much smaller than the radiation-pressure force for metals. Accordingly, we can find the steady-state velocity of the particles by setting the sum of the radiation-pressure force and the drag force equal to zero:

$$\mathbf{u} = 3 I k_{\mathrm{rp}} \mathbf{e}_z \sqrt{\frac{2\pi k T_\infty}{m}} \frac{1}{2 c p_\infty \left(8 + q\pi \sqrt{\bar{T}_r / T_\infty}\right)}. \tag{23}$$

We see from (23) that the velocity of the particle under the influence of the radiation pressure is inversely proportional to the gas pressure, in agreement with experiment [2]. Ehrenhaft [2] studied the motion of a silver sphere illuminated by focused light from an intense lamp having a broad radiation spectrum with a maximum at

$\lambda_0 = 0.7 \times 10^{-4}$ cm. Near this wavelength the complex refractive index of silver is [12] $N = 0.05 - 4i$. For estimates we assume $I = 0.84 \times 10^3$ W/cm^2, $R = 6 \times 10^{-6}$ cm, $T_\infty = 293$°K, $\varkappa_i = 4.2$ J/(cm · sec · deg), $q = 0.8$, $\gamma = 0.8$, $p_\infty = 10^6$ dyn/cm^2. Substituting these values into Eqs. (22), (13), (16), and (23), we find $k_{rp} = 0.4 \gg k_a$, $\bar{T}_S = 303$°K, $F_{rp} = 1.27 \times 10^{-11}$ dyn, and $u = 1.2 \times 10^{-3}$ cm/sec. The experimental value is $u = 2.1 \times 10^{-3}$ cm/sec. The apparent reasons for the discrepancy are the nonmonochromatic nature of the light source and the circumstance that the conditions prevailing in the experiments were $\rho \sim 1$, $R/\lambda = 1$ rather than $\rho \ll 1$, $R/\lambda \ll 1$, as we have assumed (for air, $\lambda = 6 \times 10^{-6}$ cm).

NOTATION

r, θ, φ, spherical coordinates; ω, light frequency; k_0, wave vector; λ_0, light wavelength; $N = n - i\varkappa$, complex refractive index; $J_{l+1/2}(z)$, Bessel function; $H_{l+1/2}(z)$, Hankel function; θ, angle between radius vector and light-propagation direction; $P_l^{(1)}$, associated Legendre polynomial; λ, mean free path of gas molecules; n_∞, density of gas molecules at infinity; T_∞, gas temperature at infinity; m, mass of gas molecule; q, momentum-accommodation coefficient; n_r, density of gas molecules at the surface of the particle; T_S, surface temperature; $\mathbf{n}$, unit vector normal to the surface; f^-, f^+, distribution functions of the incident and reflected molecules; $\varkappa_i$, thermal conductivity of the particle; T_i, temperature within the particle; $\mathbf{I}$, Poynting vector; I, energy flux density of radiation; R, radius of particle; k_a, absorption factor; $\mathbf{F}_p$, photophoretic force; $\mathbf{F}_d$, drag force; p_∞, gas pressure at infinity; $\mathbf{u}$, velocity of particle; $\mathbf{F}_{rp}$, radiation-pressure force; k_{rp}, radiation-pressure factor; k_c, scattering factor.

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