# RETRIEVING THE MICROSTRUCTURE OF AN ENSEMBLE OF ASPHERICAL AEROSOL PARTICLES FROM NONLINEAR OPTICAL MEASUREMENTS

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*Received May 11, 1990*

*This paper describes a technique for retrieving the size spectrum of an ensemble of aspherical particles from their preferential spatial orientation. Two basic mechanisms of particle orientation are considered. One of them has to do with aerodynamic forcing of particles moving in a viscous medium. The other stems from the interaction between the induced particle dipole moment and the electric field of the incident radiation. Quantitative estimates are presented for an ensemble of ellipsoidal particles.*

The movement of particles forced by light pressure, photophoresis and photoreactive forces in a powerful laser beam has been described in a number of papers.[1–3] However, these studies failed to notice the orienting effect of such radiation fields upon nonspherical particles.

The present paper considers several mechanisms of movement of aspherical particles immersed in a medium, resulting in their preferential spatial orientation, and comments, on the applicability of that phenomenon to the task of retrieving the aerosol microstructure from data of nonlinear optical measurements.

It is well known from hydrodynamics that a torque affects a body of asymmetrical shape in the process of its translational movement through a liquid or gaseous medium, this torque tending to orient such a body broad side the direction of movement.[4]

In particular, a rotational ellipsoid with semi-axes $a$ and $b$ ($a > b$) is forced by a torque $M$, which tends to turn its broad side perpendicular to the flow[4]

$$|M| = \frac{4}{3}\pi a^2 b u^2 \rho_m \gamma \sin\alpha, \tag{1}$$

where $u$ is the flow velocity relative to the ellipsoid; $\rho_m$ is the medium density; $\alpha$ is the angle between the direction of flow and the rotation axis of the ellipsoid; $\gamma$ is a constant.

We then obtain for a rotational ellipsoid

$$\gamma = \left[\frac{\Delta r^2}{a^2(\Delta r - bW)} - 1\right]^{-1}, \tag{2}$$

where

$$\Delta r = (a^2 - b^2)^{1/2}; \quad W = \frac{\pi}{2} - \mathrm{arctg}\,\frac{b}{\Delta r}.$$

The equation of rotational motion of the particle when that particle is affected by the torque (1) has the following form:

$$\frac{d^2\alpha}{dt^2} + W_0^2 \sin\alpha = 0, \tag{3}$$

where $W_0^2 = \frac{5}{2}\frac{\rho_c}{\rho}\frac{\gamma u^2}{ab}$; $\rho$ is the density of the material the ellipsoid is made of.

Equation (3) is the equation of a pendulum. If we assume the following initial condition: $\alpha(0) = \alpha_0$, $\frac{d\alpha}{dt} = 0$ for $\alpha = \alpha_0$, then the oscillation period, described by Eq. (3), can be calculated from the following expression[5]:

$$T = \frac{4}{W_0} F\left[\frac{\pi}{2}, k\right], \tag{4}$$

where $F\left(\frac{\pi}{2}, k\right)$ is the incomplete elliptical integral of the first kind, and $k = \sin\alpha_0/2$.

Note that

$$F\left[\frac{\pi}{2}, k\right] \simeq \frac{\pi}{2}\left[1 + \frac{1}{4}k^2 + \frac{9}{64}k^4 + \cdots\right] \approx \frac{\pi}{2},$$

i.e., it doesn't strongly depend on $\alpha_0$.

We take $\tau_{orient} = \frac{1}{4}T$ for the orientation time, since it is exactly the time required for the angle $\alpha$ to change from its initial value $\alpha_0$ to zero. The following expression is obtained for $\tau_{orient}$:

$$\tau_{\mathrm{orient}} = \frac{\pi}{2u}\left[\frac{2\rho ab}{5\rho_m\gamma}\right]^{1/2}. \tag{5}$$

The random movement of the aerosol particles in the medium results from their Brownian motion and from the eddy diffusion of the medium. While the velocity Brownian motion for a given particle depends

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on the medium temperature alone, the eddy mixing intensity in air j, being a measure of the pulsation component of the wind velocity varies over a wide range (from 0.002 to 0.20) in the atmosphere.[6]

The particle acquires some directional movement with respect to the medium due to the gravitational force and the light pressure.[1] Also the radiative and light reaction forces affect the particle in a laser radiation field.[2]

The particle velocity due to the gravitational force in a viscous medium exceeds the rms pulsation velocity of the average wind velocity $u_w$ for particles of size

$$r > \left[ \frac{j u_w 9\eta}{2g(\rho - \rho_m)} \right]^{1/2}, \tag{6}$$

where $g$ is the free-fall acceleration; η is the medium dynamic viscosity.

For $u_w = 1$ m/s, $j = 0.1$, $\rho = 10^3$ kg/m$^3$, $\rho_m = 1.2$ kg/m$^2$, $\eta = 1.8 \cdot 10^{-5}$ kg/(m · s), and $g = 10$ m/s, $r > 28$ μm.

Thus particles of arbitrary shape but of an effective size of several dozen microns begin to move directionally through the atmosphere, and because of aerodynamic forcing should orient with their maximum cross section facing the direction of their movement. This effect is indeed observed for falling snowflakes and ice crystals.[7,8]

A particle, forced by light pressure, reaches a certain stationary velocity $u$, which may be easily computed from the Stokes equation. The velocity $u$ exceeds the Brownian velocity of the particle if the power density $J$ of the forcing radiation exceeds a certain threshold value, given by the expression[9]

$$J_{thr} = \frac{9c\eta}{r^2 K_{LP}} \left( \frac{kT}{\pi r \rho} \right)^{1/2} \left[ 1 - \exp\left( - \frac{9\eta\tau_{pulse}}{2r^2\rho} \right) \right]^{-1}, \tag{7}$$

where $K_{LP}$ is the light pressure cross section of the particle, $k$ is the Boltzmann constant, $T$ is the medium temperature, $c$ is the speed of light, $\tau_{pulse}$ is the duration of the laser pulse.

For the equation of a pendulum and relation (5) to be applicable to the calculations of the particle orientation time in a laser beam (due to aerodynamic forcing) the time the particle takes to reach its stationary velocity $\tau_{stat}$ should be much less than that calculated using Eq. (5).

It can be shown that $\tau_{stat} = \dfrac{2r^2\rho}{9\eta}$, i.e., it depends on the properties of the particle and the medium.

Calculations show that the equation of a pendulum is applicable to aerosol particles with $r < 5$ μm in air if their density is $\rho \simeq 10^3$ kg/m$^3$. The process of orientation for coarse aerosol particles is described by much more complex differential equations.

For particles with $r < 5$ μm suspended in air we obtain following expression for their orientation time:

$$\tau_{orient} = \frac{3\pi c\eta}{K_{LP}J} \left[ \frac{2}{5} \frac{\rho}{\rho_m} \frac{1}{\gamma\zeta^{1/3}} \right], \tag{8}$$

where $\zeta = a/b$ is their asphericity parameter.

Figure 1 shows a graph of the orientation time vs the effective size of the rotational ellipsoid for $\zeta = 2$, 3, 5. The radiation power density was taken to be $J = 5 \cdot 10^5$ W/cm$^2$ at $\lambda = 10.6$ μm. It is important to note that the orientation time is observed to increase as the effective size of the aspherical particles decreases. This effect is due to a decrease in the light pressure cross section for smaller particles.[10]

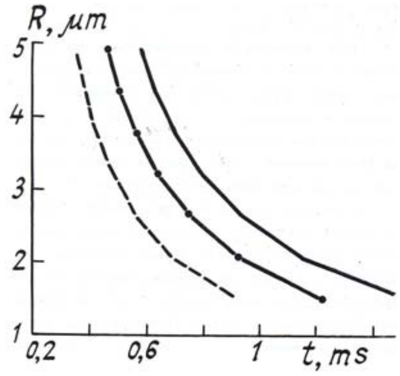


*FIG. 1. Orientation time for three types of ellipsoids: $\zeta = 2$ (solid line), $\zeta = 5$ (marked line), $\zeta = 5$ (dashed line), when there is the aerodynamic effect. The radiation parameters:* $\lambda = 10.6$ μm *and* $J = 5 \cdot 10^5$ W/cm$^2$.

Another particle-orienting mechanism plays an important role. Along with the orientation of aspherical particles as a result of aerodynamic forcing by the moving medium, such particles may be oriented in an external homogeneous electric field. The cause of such a phenomenon lies in the interaction between the dipole moment of a particle polarized in an external field and the field itself. The torque $M$ acting upon a rotational ellipsoid situated in a homogeneous electric field $E$ is equal to[11]

$$|M| = \frac{(\varepsilon - 1)^2 \cdot |1 - 3n| \cdot V \sin 2\beta}{8\pi(n\varepsilon + 1 - n)[(1 - n)\varepsilon + 1 + n]} E^2, \tag{9}$$

where β is the angle between the field $E$ and the ellipsoid symmetry axis, ε is the dielectric constant of the ellipsoid, $n$ is the depolarization coefficient along the ellipsoid axis, and $V$ is the ellipsoid volume.

The depolarization coefficient $n$ depends on the ellipsoidal shape alone, and its numerical values for various ellipsoidal shapes may be found in Ref. 12. The total torque $M$ is oriented so as to turn the prolate axis of the ellipsoid ($n < 1/3$) parallel to the field, and that of the oblate one ($n > 1/3$) — perpendicular to that field.

Carrying out similar operations with the solution of the equation of a pendulum, with the torque (9), we obtain, as before, the following expression for the particle orientation time:

$$t = \pi/W_1, \tag{10}$$

where

$$W_1 = \left[ \frac{5(\varepsilon - 1)^2 |1 - 3n| E^2}{8\rho ab\pi(n\varepsilon + 1 - n)[(1 - n)\varepsilon + 1 + n]} \right]^{1/2}.$$

The calculations yield an orientation time $t = 3.8 \cdot 10^{-2}$ s for $E = 10^2$ V/m, $\varepsilon = 20$, $\rho = 10^3$ kg/m$^3$, $r = 5$ μm.

If the fields vary with time and the particle size is much less than the radiation wavelength, the orientation time for such particles may be described by expression (10), but the frequency dependence of the particle dielectric constant must then be taken into account, and $\langle E^2 \rangle$ (the squared field average over the oscillation period) must be substituted into that expression in place of $E^2$.

Figure 2 shows the dependence of such an orientation time for an ellipsoid with semi-axes ratio $a/b = 2$ on the effective size of the ellipsoid for radiation power density $J = 5 \cdot 10^9$ W/m$^2$.

Note that the orientation time of the particle due to aerodynamic forcing $\tau_{orient} \sim 1/J$, while that due to the external electric field $t \sim 1/\sqrt{J}$, so that these orientation times become comparable at a radiation power density of

$$J = \frac{90\eta^2 c(\varepsilon - 1)^2 |1 - 3n|}{\gamma\rho_m r^2 \varepsilon^{-1} K_{LP}^2 (n\varepsilon + 1 - n)[(1 - n)\varepsilon + 1 + n]\sqrt{\varepsilon}}. \tag{11}$$

Calculations show that $J = 10^{11}$ W/m$^2$ for $\eta = 1.8 \cdot 10^{-5}$ kg/(m · s), $r = 2$ μm, $n = 0.25$, $\varepsilon = 2$, $\rho_m = 1.3$ kg/m$^3$, and $K_{LP} = 0.1$.

Hence the torque due to aerodynamic forcing may be neglected, compared to that from the external electric field, for every conceivable radiation power density. Note that a similar situation is also observed in other, more viscous media, e.g., water.

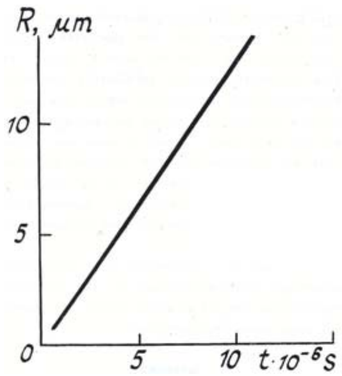


*FIG. 2. Orientation time for electric dipole interaction for an ellipsoid with $\zeta = 2$. Radiation power density $J = 5 \cdot 10^{-5}$ W/cm$^2$.*

It should be recalled that expression (10) is valid only for particles of size much less than the forcing radiation wavelength $r \ll \lambda$.

The cases $r \sim \lambda$ and $r \gg \lambda$ are not considered in our study.

The phenomenon of particle orientation in a laser radiation beam allows the possibility of retrieving the aspherical particle size spectrum from the observed dynamics of the aerosol extinction.

Not allowing for the dependence of the particle scattering properties on the polarization state of the radiation, an expression which describes the time dependence of the aerosol extinction coefficient reads

$$\Delta\beta(t) = \int_{r_1}^{r(t)} \left[K_R(r, m) - K_N(r, m)\right] r^2 F(r) dr, \tag{12}$$

where $K_R(r, m)$ is the efficiency factor for the randomly oriented particles, $K_N(r, m)$ is that factor for particles oriented with their broad face perpendicular to the laser beam; $F(r)$ is the size distribution of these particles; $r(t)$ is the size of the particles that have already been oriented as of the time $t$; $r_1$ is the particle minimal size.

Differentiating Eq. (12) with respect to time, we obtain

$$F(r) = \frac{\Delta\beta'(t)}{r'(t)} \frac{1}{\left[K_R(r, m) - K_N(r, m)\right] r^2} \Bigg|_{r=r(t)} = \frac{\Delta\beta'(t)}{\varphi(t)}. \tag{13}$$

The calculational data on the extinction efficiency factor for both the prolate and the three oblate rotational ellipsoids ($\zeta = 2, 3, 5$) for $m = 1.33$ are presented in Refs. 13 and 14.

Figure 3 demonstrates the dependence of $1/\varphi(t)$ for an ellipsoid with $\zeta = 5$ for three different radiation power densities: $10^5$, $5 \cdot 10^5$, and $4 \cdot 10^4$ W/cm$^Z$ ($\lambda = 10.6$ μm), obtained taking relation (10) into account.

To test algorithm (13) for $F(r)$ both the direct and inverse problems were solved for a set of spheroids with $\zeta = 5$, $m = 1.33$, and $\rho = 1$ g/cm$^3$ within the size range 1.5–14 μm, described by the distribution function

$$F(r) = dr_m^{k_1} \cdot \exp\left[-\beta'' r^{k_2}\right], \tag{14}$$

where $d$, $k_1$, $k_2$, and $\beta''$ are fixed constants, and $r_m$ is the modal radius.

The particles were assumed to be irradiated at a power density of $4 \cdot 10^4$ W/cm$^2$ and to be sensed by a weak beam with $\lambda = 5$ μm. It was also assumed that the particles orient as a result of forcing by the external electric field alone. We used the single-parameter distribution function (14), which does not account for the particles' initial orientation (i.e., for the angle $\alpha_0$ in Eq. (4)). Such a simplification is permissible because the orientation time depends only weakly on $\alpha_0$, as can be seen from Eq. (4), and we can take $\alpha_0 = \pi/4$ for all the particles.

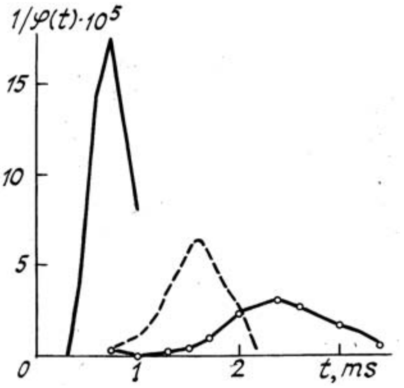


*FIG. 3. The function $\varphi(t)$ for ellipsoids with $\zeta = 5$ at three different radiation power densities: $J = 10^5 W/cm^2$ (solid line), $J = 5 \cdot 10^5 W/cm^2$ (dashed line), and $J = 4 \cdot 10^5 W/cm^2$ (marked line).*

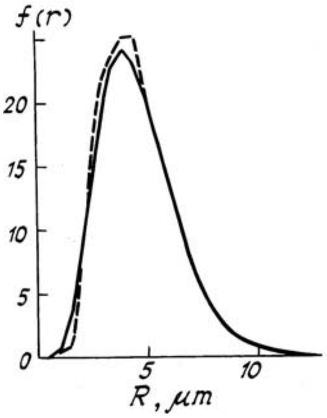


*FIG. 4. Actual (solid line) and calculated (dashed line) particle size spectra.*

Figure 4 shows the "actual" (14) and computed (13) distribution functions, obtained for $d = 2.373$, $k_1 = 6$, $\beta'' = 3/2$, and $k_2 = 1$. They are in satisfactory agreement with each other. The calculations were performed on an ES-1066 computer.

Although our calculations cover only certain prescribed rotational ellipsoids, the above-noted regularities can, to a certain extent, be found in all other aspherical particles.

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