Study of particle shapes by the light-beating method

A. V. Lomakin and V. A. Noskin

B. P. Konstantinov Institute of Nuclear Physics, USSR Academy of Sciences

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It is shown theoretically and experimentally that for anisotropic particles placed in a laminar stream with a transverse velocity gradient, the self-beating spectrum will contain split-off lines, and analysis of the positions and shapes of these lines yields information on the anisotropy of these particles and on their dimension distribution.

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Macroparticles in solutions are very frequently investigated by the light beating spectroscopy technique.[1] Its advantage over traditional gasdynamics, sedimentation, optical, and other methods are that it yields rapidly and with high accuracy the same

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information on the system, without perturbing the system at all.

The gist of the method is the following. The investigated sample is illuminated with monochromatic light from a radio. The different motions of the particles in the solution (translational and rotational diffusion, motion under the influence of external fields, chemical reactions, etc.) change the spectral properties of the incident light. By determining these changes with high accuracy, i.e., by measuring the difference effect, it is possible to assess the behavior of the particles in the solution. Since we are dealing with relatively large objects (larger than 100 Å), which produce small changes in the spectral properties of the incident light ($\Delta\lambda/\lambda \sim 10^{-14}$–$10^{-11}$), it becomes possible to analyze the effect by modern high-power radiotechnical methods. These include, in particular, real-time spectrometers.[2]

The use of this procedure is particularly promising for systems that are not in thermodynamic equilibrium.[3] By determining the "response" of the system to an external perturbation it is possible to expand greatly the information on its kinetic characteristics, as well as on the distribution of the investigated particles with respect to dimensions or other parameters. It is quite obvious that a successful choice of the perturbing field is of decisive importance here.

This is why interest attaches to the case considered in the present paper, namely the onset of dynamic effects in the field of hydrodynamic forces of a stream with a transverse velocity gradient. This problem is closest in its concept to the dynamo-optical Maxwell effect, the only difference being that in addition to the orienting action of the stream we are interested in the dynamic behavior of asymmetrical particles in the stream, which yields more information than the widely used technique of birefringence in the stream.[4]

The idea of the method is the following: the particle in a stream with velocity gradient is in the state of rotational motion; its form factor then undergoes periodic modulation and this, as will be shown in this paper, produces in the scattered-light spectrum lines that are split off by an amount equal to the frequency of this rotation. The problem is considered by us theoretically and illustrated with experiments. The most interesting results of the theoretical investigations are the following: a) equations of motion have been obtained for a large class of objects—rigid axially-symmetrical particles; b) the period of motion of the particle axes was determined and turned out to be independent of the initial orientation of the axes. The latter circumstance is particularly important, for otherwise, after averaging over the particle orientations, the effect would turn out to be "smeared out."

We describe briefly the different stages of the theoretical analysis. Let a particle with an axis whose direction is specified by a unit vector n_i be situated in a liquid stream with a velocity gradient w_{ik} and have an instantaneous angular velocity Ω_i. Strictly speaking, flow around a rotating anisotropic body is not stationary but it can be shown that in a laminar stream (Reynolds number $\mathrm{Re} \ll 1$) the effects connected with the nonstationarity as well as with the inertia of the particles are small in terms of the parameter R. If we neglect these effects, then the angular velocity, naturally, is proportional to w_{ik} and is determined only by the instantaneous position of the axes:

$$\Omega_i = D_{ikl}(\mathbf{n})\, w_{kl}\,, \qquad (1)$$

To determine the structure of the tensor D_{ikl} we note that it connects the axial vector Ω_i and the polar tensor w_{kl}, and therefore contains a fully antisymmetrical unit tensor ϵ_{ikl}. This condition, together with the requirement that Eq. (1) be invariant to a transition to a rotating coordinate system, enables us to determine D_{ikl}:

$$D_{ikl} = \frac{1}{2}\epsilon_{ikl} + \frac{a}{2}\left(\epsilon_{ikm}n_m n_e + \epsilon_{ilm}n_m n_k\right). \tag{2}$$

Substituting (1) and (2) in the equation of motion of the particle axes

$$\frac{dn_i}{dt} = \epsilon_{ikl}\,\Omega_k n_e, \tag{3}$$

we can determine the time dependence of $n_i(t)$, which turns out to be periodic with a period

$$T = \frac{4\pi}{w\sqrt{1-a^2}}, \tag{4}$$

regardless of the initial conditions. (We note that for a particle with lower symmetry the motion is no longer periodic at all.)

The parameter a in Eq. (2) characterizes the anisotropy of the particle. It follows from the analysis of the equations of motion that $-1 < a < 1$($a = -1$ for an infinitely long cylinder, $a = 1$ for an infinitely thin disk: $a = 0$ for a sphere). Calculation of the anisotropy parameter calls for a complete solution of the hydrodynamic problem of flow with velocity gradient around a body, which in the general case is an exceedingly complicated problem. In the particular case of an ellipsoid of revolution, however, it is connected with the axis ratio p by the formula

$$a = \frac{1-p^2}{1+p^2}. \tag{5}$$

The object of the investigation was suspension of the bacteria *Escherichia coli*, whose dimensions (length 2 μm and more, diameter 0.8 μm) ensured an appreciable magnitude of the expected effect. The experimental results, which confirm the basic conclusions of the theoretical analysis, are shown in Fig. 1, where, in addition, the optical part of the installation is shown schematically. The electronic part of the installation did not differ from that described by us earlier.[2]

Curve 1 of the diagram shows clearly the split-off line and, if a more careful examination is made, also the next harmonic. The splitting is ~ 120 Hz. In our experiment the frequency of rotation of the rotor of radius $R = 18$ mm was $f = 36$ Hz. At a gap $d = 2$ mm, this corresponded to velocity gradients $w = 2\pi fR/d \sim 2\times 10^3$ sec^{-1}. Using (4) and (5) we can find the ratio of the axes of the equivalent ellipsoid. The obtained value $p \sim 2.3$ is in good agreement with that observed visually.

For comparison, Fig. 1 shows data for the same solution with the rotor immobile (curve 2) and in the presence of a velocity gradient, but for spherical particles (latex of ~ 1 μm diameter, curve 3). The absence of an effect in the latter case is due to the absence of modulation of the form factor. Comparing curves 1 and 2 we see that the width of the split-off peak (curve 1) exceeds the diffusion width (2), in complete correlation with the polydisperse character of the chosen object.

We indicate in conclusion that the main factor that determines the limits of

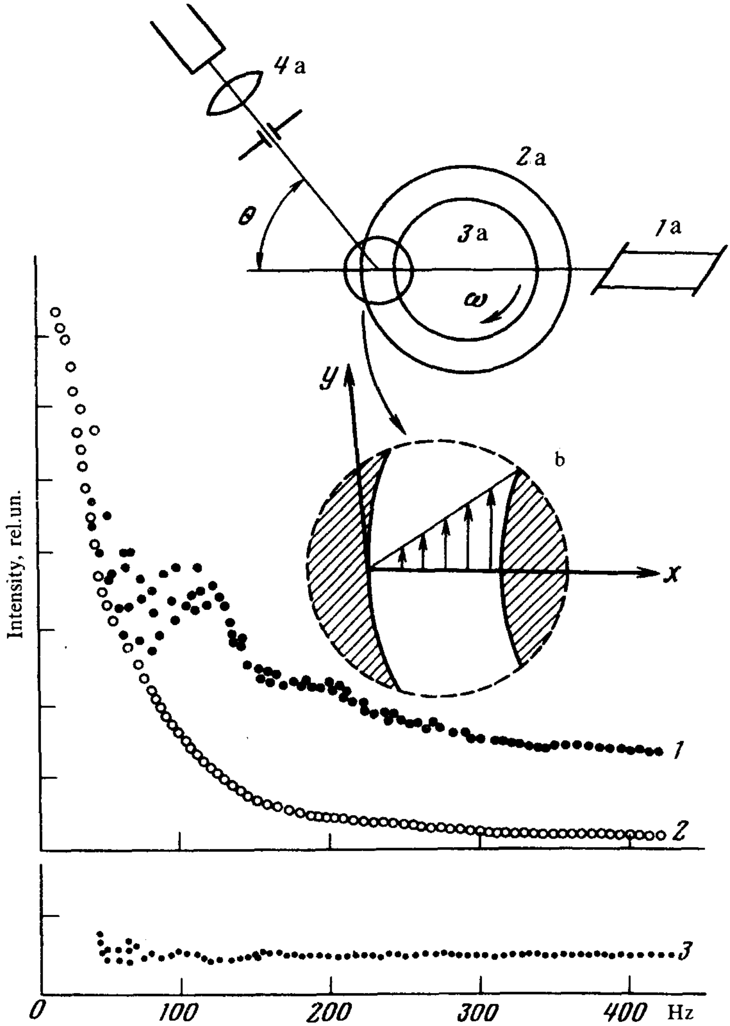

FIG. 1. Optical system of experiment and the obtained spectrograms for three cases: 1) solution of *E. coli* in the presence of a velocity gradient; 2) solution of *E. Coli* with the rotor stationary; 3) solution of latex particles with $d \sim 1$ μm in the presence of a velocity gradient. In all cases the scattering angle is $\theta = 90°$, the scanned band is 500 Hz, the spectral resolution was 2.5 Hz. In the diagram: 1a) LG-38 laser; 2a and 3a) stator and rotor; the gap between them is filled with the investigated substance; 4a) photomultiplier with lens and collimator, b) Pattern of the velocities in the cell gap.

applicability of this method is the particle dimension; if the particle dimensions are too small compared with the wavelength of the light, the effective form factor modulation will not be noticed. A theoretical estimate yields for the minimum necessary particle dimension a value on the order of 100 Å.

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