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ФІЗИКА АЕРОЗОЛІВ

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Study of the Disperse Composition of Suspensions and Sputtered Substances by means of Small-Angle Light Scattering

Spatial distribution of the light scattered by a disperse system of particles depends on their sizes, shapes, positions, etc., which can be used for experimental determination of the parameters mentioned. For stochastic systems with the particles' sizes exceeding the radiation wavelength, most of the scattered radiation concentrates near the incident beam axis. In this small-angle approximation, the scattering pattern is especially simple and regular, which enables to develop efficient procedures for the disperse system investigation. We describe the algorithm for determination of the mean particle radius in the system with lognormal distribution of the particle sizes and negligible multiple scattering. The algorithm's performance is demonstrated on the practical example of the "fog" generated by a gasoline injector. The ways are discussed for further algorithm generalization and its extension to a non-parametric analysis of disperse systems with a priori unknown form of the particle sizes' distribution.
Keywords: *disperse system, light scattering, particle size, distribution function, experimental measurement*

Introduction. Optical methods for studying the structure of various substances and physical and chemical processes occurring in them have been used for many years and have repeatedly proved their effectiveness. Due to the intensive development of optical and laser technologies, numerous new possibilities and applications have emerged in this direction. Optical methods for studying disperse systems based on the analysis of the characteristics of electromagnetic radiation scattered by a system of small particles differing in size, shape, and physical properties appear to be especially promising and sometimes irreplaceable [1].

A general scheme of optical investigation of disperse systems is as follows [2] (see Fig. 1). A light beam with known characteristics (most often it is a plane wave or a Gaussian laser beam) is incident on the system. Let the incident light intensity equal to I_0 and is the same for all particles composing the system (this is a good approximation if the incident beam radius exceeds the system total size). Part of the incident light passes through the system without interacting with particles and forms a beam of the same spatial structure as the initial one, but somewhat weakened in intensity I_1; the ratio I_1/I_0 characterizes the system's extinction. Another part of the incident light is absorbed in the system; it characterizes the energy loss in the system. But the most important is that part of the light that interacts with the system and, afterwards, di-

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verges in various directions. This is scattered light, characteristics of which depend on the properties of the scattering system: the size and dimensions of the particles, their physical and chemical nature, shape, etc. Therefore, by studying the characteristics of the scattered light, one can also learn the properties of the scattering system [2].

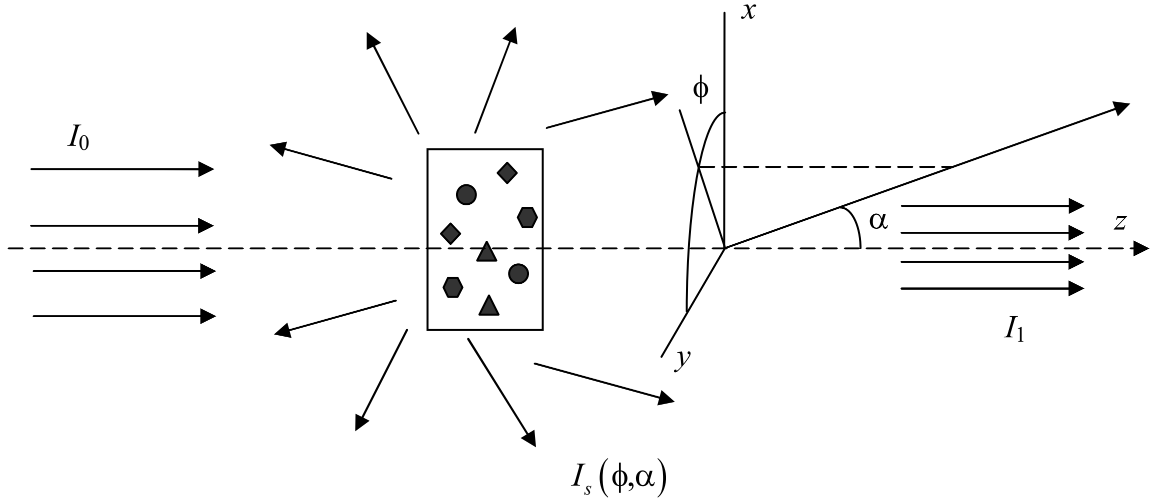

Fig. 1. General illustration of the light scattering by a disperse system

Formulation of the problem. The most important characteristic of the scattered light is the scattering indicatrix – a function that describes the distribution of scattered radiation over the angles of a spherical coordinate system ϕ and α (Fig. 1). Let the scattering indicatrix for a single particle be $u(\phi, \alpha, b_1, b_2, \ldots, b_n)$, where $b_1, b_2, \ldots, b_n$ are parameters that characterize, in particular, the size, shape, and optical properties of the particle. With assumption of a low particle density, which allows one to neglect the effects of multiple scattering, and supposing a random arrangement of particles in the system, we can assume that the scattering from a set of particles is determined by the sum of contributions from each particle separately [2, 3]. If the particles are not the same, then the system can be characterized by the distribution function $f(b_1, b_2, \ldots, b_n)$ of the particles with respect to the parameters $b_1, b_2, \ldots, b_n$, satisfying the normalization condition

$$\int f\left(b_1, b_2, \ldots, b_n\right) db_1 db_2 \ldots db_n = 1\,. \qquad (1)$$

In this case, the intensity of light scattered by the system of particles in the direction specified by angles ϕ, α is determined by the equation

$$I_s\left(\phi,\alpha\right) = N\int u\left(\phi,\alpha,b_1,b_2,\ldots,b_n\right) f\left(b_1,b_2,\ldots,b_n\right) db_1 db_2 \ldots db_n\,. \qquad (2)$$

where N is the total number of particles participating in the scattering.

Equation (2) is the basic equation of the problem under consideration. On the one hand, it allows, knowing the form of the function $u(\phi, \alpha, b_1, b_2, \ldots, b_n)$, which is determined theoretically depending on the nature and shape of particles, and the distribution function $f(b_1, b_2, \ldots, b_n)$, to calculate the angular distribution $I_s\left(\phi,\alpha\right)$ of the radiation scattered by the system, i.e. to solve the direct scattering problem. On the other hand, it shows that, having measured $I_s(\phi,\alpha)$ experimentally, one can find the function $f(b_1, b_2, \ldots, b_n)$ and, thus, determine the important characteristics of the system and its constituent particles. This operation will solve the inverse scattering problem.

As can be seen from (2), the solution of the inverse problem requires the solution of an integral equation. Therefore, this problem is generally more complex and its solution is not always possible. In this case, various simplifying assumptions arising from a specific experimental situation can be of great help.

Let us consider the important case when the set of the particles' parameters $\{b_1, b_2, \ldots, b_n\}$ consists of a single parameter – the particle radius $b_1 \equiv a$. Then, if the condition $a \gg \lambda$ is satisfied, where λ is the radiation wavelength, the function $I_s(\phi,\alpha)$ does not depend on the azimuthal angle ϕ and differs significantly from zero only at small ϕ: the situation of small-angle scattering is realized [4–8]. In this case, scattering by a single particle satisfies the conditions of Fraunhofer diffraction and is described by the function [4]

$$u(\alpha,a)=\left[2\pi a\frac{J_1(k\alpha a)}{k\alpha}\right]^2, \tag{3}$$

where $k = 2\pi/\lambda$ is the radiation wave number, J_1 is the notation of the Bessel function [9]. This function is maximal at $\alpha = 0$ and has an infinite number of zeros whose positions are determined by the particle size. Practically important are the zeros situated closest to the axis z ($\alpha = 0$), the first of which equals to $\alpha = 3.8/(ka)$.

If all particles of the system have the same size, then the distribution of the scattered radiation over the angle α $I_s(\alpha)$ is just proportional to expression (3). However, it is much more common for the particles to be of different sizes; in aerosol systems, the particle size distribution is usually described by the lognormal distribution function [2–4]

$$f(a)=\frac{1}{a\sigma\sqrt{2\pi}}\exp\left[-\frac{1}{2}\left(\frac{\ln a-\mu}{\sigma}\right)^2\right]. \tag{4}$$

Here, the distribution parameters are present: σ characterizes the dispersion of particles in size, so that at $\sigma = 0$ the distribution is monodisperse, and μ determines the average particle size a_c in accordance with the equality $a_c = \exp(\mu + \sigma^2/2)$. From equation (2), with allowance for (3) and (4), one obtains

$$I_s(\alpha)=\frac{(2\pi)^{3/2}}{\sigma(k\alpha)^2}\int_0^\infty a\exp\left[-\frac{1}{2}\left(\frac{\ln a-\mu}{\sigma}\right)^2\right]J_1^2(k\alpha a)\,da. \tag{5}$$

Numerical algorithm and solution. The algorithm for determining the dispersion parameters (in the simplest case, the average size a_c) is based on comparing the experimentally measured integral scattering pattern with the theoretically calculated one (see Fig. 2). In the experimental modeling of the small-angle scattering situation we used the gasoline injector producing a nearly monodisperse "fog" of spherical particles. In general features, the experimental procedure reproduces the scheme of Fig. 1: a transparent cell containing the suspension was illuminated by the collimated beam of a semiconductor laser (λ = 0.67 μm), the near-axis scattered radiation is collected by a focusing lens (not shown in Fig. 1), and the focal-plane pattern was registered by a CCD web-camera (Fig. 3a). The non-scattered part of the incident beam with the intensity I_1 (see Fig. 1) was stopped by an opaque screen whose shadow is seen in the center of Fig. 3a.

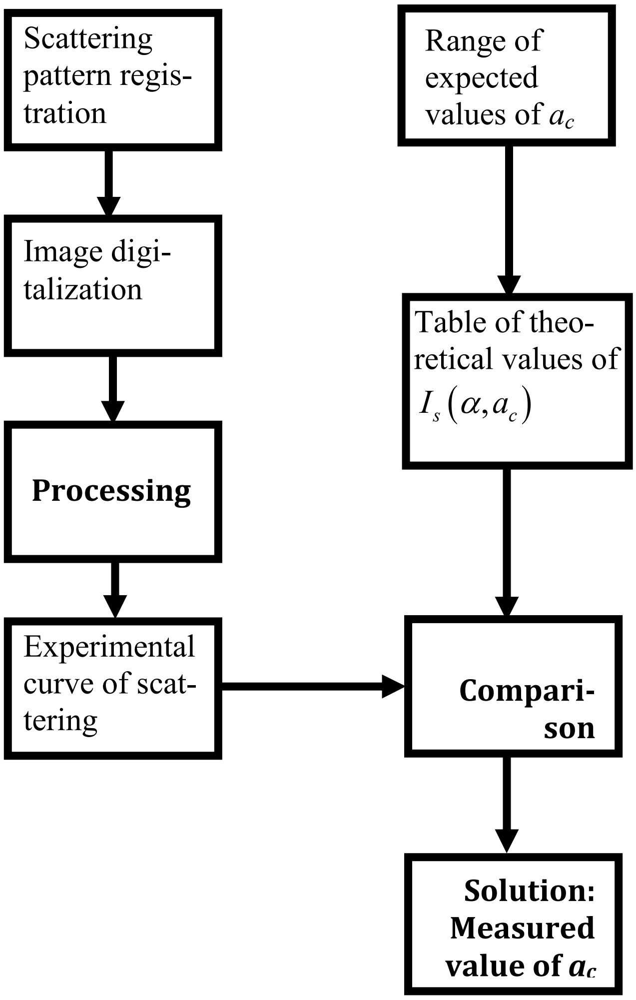

Fig. 2. Scheme of the algorithm for measurement of the disperse system characteristics

The dependence of the scattered power on the scattering angle α is proportional to the illumination brightness dependence on the polar radius, which is calculated by averaging the observed intensity inside the rings of a given radius (shown by thin black circular contours in Fig. 3a). The resulting experimental curve (red in Fig. 3c) is compared with a set of pre-calculated theoretical curves determined by equation (5) for different values of the average particle radius; the variance parameter was set as $\sigma = 0.01$. The value of a_c at which the best approximation is observed (estimated by the least square method) is taken as the real value of the average radius of particles in the studied system (in the example of Fig. 3, $a_c \approx 1.5$ μm). Estimation of additional distribution parameters (variance, etc.) can be performed according to the same scheme.

Discussion and conclusion. The rather good agreement between the experimental and theoretical curves in Fig. 3c testifies for the validity of the approximations presumed, in particular, of the log-normal particle-size distribution (4). In many practical cases it is impossible to tell in advance that the particle size distribution function has a certain form. Then, a nonparametric determination of the distribution function is also possible, i.e. direct finding of a discrete set of the function $f(a)$ values. For example, we again restrict ourselves to the case of only one parameter – the particle radius a. In this situation, the integral equation (2) can be written in the form

$$I_s(\alpha) = \int u(\alpha, a) f(a) da \,. \tag{6}$$

and, after transformation to the numerical form, reduces to the system of linear equations

$$\mathbf{I} = \mathbf{Af}\,, \tag{7}$$

where

$$\mathbf{I} = \begin{pmatrix} I(\alpha_1) \\ I(\alpha_2) \\ \vdots \\ I(\alpha_n) \end{pmatrix}$$

is a vector of the measured values of the intensity scattered in directions specified by the polar angles α_1, α_2, … α_n, and

$$\mathbf{f} = \begin{pmatrix} f(a_1) \\ f(a_2) \\ \vdots \\ f(a_m) \end{pmatrix}$$

is the sought vector of the distribution function values. The matrix A is composed by the theoretically calculated values of the scattering indicatrix $u(\alpha_i, a_i)$ (e.g., those described by (3) in the small-angle scattering situations) for equidistant values of a_1, a_2, … a_m:

$$\mathsf{A} = \Delta a \begin{pmatrix} u(\alpha_1, a_1) & u(\alpha_1, a_2) & \dots & u(\alpha_1, a_m) \\ u(\alpha_2, a_1) & u(\alpha_2, a_2) & \dots & u(\alpha_2, a_m) \\ \vdots & \vdots & \vdots & \vdots \\ u(\alpha_n, a_1) & u(\alpha_n, a_2) & \dots & u(\alpha_n, a_m) \end{pmatrix}$$

and $\Delta a = a_{i+1} - a_i$ is the size of the intervals into which the integration domain of (6) is divided. Solution of the system (7) is possible if $n \geq m$; when $n = m$ this is a simple system of linear equations, but if $n > m$ the system of equations (5) determines the vector **f** statistically by the least-square method:

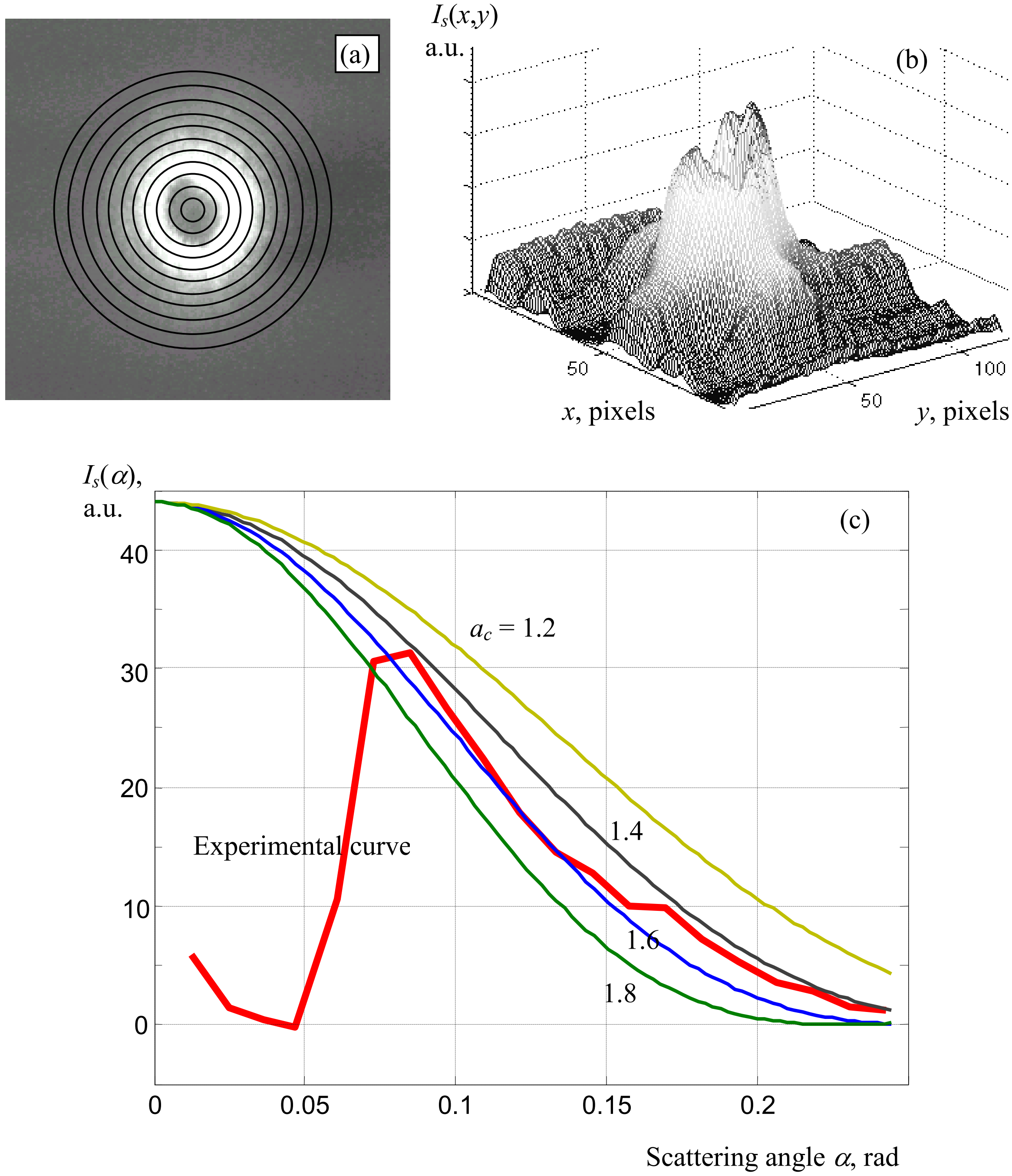

Fig. 3. (a) Scattering pattern registered by the camera (the central dark spot is formed by the opaque “stopper” of the probing beam I_1); (b) 3D intensity plot of the digitalized image (a); (c) Experimental angular intensity distribution (red) and illustration of the adjusting procedure (colored lines are calculated for different values of the parameter a_c indicated near each curve).

$$\mathbf{f}=\left(\tilde{A}A\right)^{-1}\tilde{A}\mathbf{L}.$$

Thus, the solution of the inverse scattering problem is achieved. The greater the difference $n-m$, the more probable is the stability and regularity of the solution. To increase the stability, additional regularization procedures can be employed based on a comparative reliability assessment for various elements of the experimental data vector **I** [10].

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Дослідження дисперсного складу суспензій та розпорошених речовин методом малокутового розсіювання світла

АНОТАЦІЯ

Просторовий розподіл світла, розсіяного дисперсною системою частинок, залежить від їх розмірів, форми, розташування тощо, що може бути використано для експериментального визначення вказаних параметрів. Для стохастичних систем з розмірами частинок, що перевищують довжину хвилі випромінювання, більша частина розсіяного світла концентрується поблизу осі зондуючого пучка. У цьому малокутовому наближенні картина розсіювання є особливо простою та регулярною, що дозволяє розробити ефективні процедури для дослідження параметрів дисперсної системи. Описано алгоритм визначення середнього радіуса частинок у системі з логнормальним розподілом частинок за розмірами в умовах нехтовно малого багатократного розсіювання. Ефективність алгоритму продемонстрована на практичному прикладі "туману", який утворюється форсункою для розпилювання рідкого палива. Обговорюються шляхи подальшого вдосконалення алгоритму та його поширення на непараметричний аналіз дисперсних систем з апріорно невідомою формою розподіла частинок за розмірами.

Ключові слова: *дисперсна система, розсіювання світла, розмір частинок, функція розподілу, експериментальне визначення.*