SIMULATION OF AN EXPERIMENTAL SETUP FOR MEASUREMENT OF LIGHT SCATTERING BY POROUS FILMS

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A mathematical model of an experimental setup for light scattering measurements has been developed. This model allows one to reconstruct the intensity distribution of scattered light, which is not distorted by an measuring setup. The application of the approximating "hat" function as an apparatus function has been justified.

Introduction

In our experiments, the measurements of weak signals from scattered light were conducted with a wide-aperture diaphragm on the registering device in order to enhance the signal-to-noise ratio. The wide aperture of the diaphragm leads to considerable losses in the precision of the data obtained whereas it is necessary to achieve a high angular resolution of the signal, as in the case of spectrometric researches or the study of scattering processes. Knowledge of the apparatus function of a measuring setup allows one to enhance its angular resolution.

The idea of this work was to obtain the apparatus function of the setup for light scattering measurements and an analytic approximation of this function.

In the experiments, the indicatrix of light scattering by nanoporous films (in particular, titanium dioxide films) has been measured. One of the tasks of conducted measurements was to obtain a true angular distribution of the scattered light intensity, i.e., the distribution which is not distorted by a measuring setup. In order to solve this problem, one can use several approaches. The first approach is to use a perfect measuring device with half-width of the apparatus function which is considerably narrower than details of the scattered light distribution. The second one is to apply the methods of mathematical interpretation for the analysis of experimental results.

The essence of the mathematical interpretation is that, in most physical researches, the sought element v which characterizes an investigated physical object, a phenomenon, or their properties, cannot be observed directly, and therefore we investigate its certain manifestation which can be described as follows:

$$u = Av, \quad A: V \to U, \quad v \in V, \quad u \in U.$$

That is, some image u of the actual state v of an object, a phenomenon, or a process is recorded.

When solving the problem of reconstruction of the element v, one should make distinction between two main stages [1]:

— construction of an operator A acting on the element v; $v \in V$, $u \in U$, $A: V \to U$;

— determination of the element v via an element u and the operator A, i.e., the solution of the operator equation $Av = u$; $v \in V$, $u \in U$. The construction of the operator A is known as the modelling stage of the measuring tract of an experimental setup, since just the measuring tract distorts the element v. The operator A can be given as [2]

$$A\,v \equiv \int_a^b K(x,s)\,v(s)ds, \tag{1}$$

where $K(x,s)$ is an apparatus function such that

$$\int_a^b K(x,s)v(s)\,ds = u(x), \quad c \le x \le d,$$

$$K(x,s) \in C([c,d] \times [a,b]), \quad u \in U, \quad U = L_2(c,d). \tag{2}$$

In physical researches, an apparatus function is the characteristics of a linear measuring device and sets a relation between a measured magnitude at the output of the device with an actual magnitude at its input [3].

According to this approach, the problem is reduced to the determination of the function $v(x)$ through the

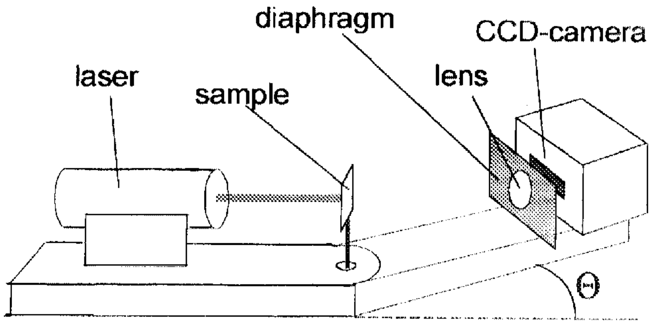

Fig. 1. Experimental setup

known apparatus function $K(x,s)$ and measured function $u(x)$, i.e. to the solution of a first kind Fredholm linear integral equation which has, in the general case, a representation

$$u(x) = \int_a^b K(x,s)\, v(s)\, ds. \tag{3}$$

The scheme of an experimental setup on the basis of a goniometer G-5 is shown in Fig. 1. We used a He-Ne laser ($\lambda = 632.8$ nm; with a power of 1 mW) with Gauss distribution of the intensity and a characteristic beam diameter of 0.95 mm in the specimen plane. Specimens were mounted on the goniometer axis perpendicularly to the laser beam. To collect the scattered light, we used a 1024-pixel (in line) CCD-camera with a diaphragm and a focusing lens before it which were fixed on a moving arm of the goniometer at a distance of 8.6 cm from the specimen. We have measured the dependence of the scattered light intensity on a polar angle θ of a spherical reference system in the plane $\varphi = 0$ (the axis $\theta = 0$ is directed along the laser beam) for the samples of nanoporous TiO_2 films on a glass substrate with thickness about 1 micron. The scattered light intensity $I(\theta)$ was determined as a sum of intensities at all elements of the CCD-camera.

A mathematical interpretation of the experimental results means, first of all, the derivation of information by using a model experimental setup, whose characteristics may exceed the limiting attainable ones of a real experimental setup. Therefore, the solution of the problem of enhancement of the experimental setup capabilities starts from the construction of a mathematical model for the setup.

The question about a relationship between the scattered light intensity before the diaphragm and the intensity registered by the CCD-camera arises during the analysis of experimental results. An answer can be given in the following manner (see Fig. 2).

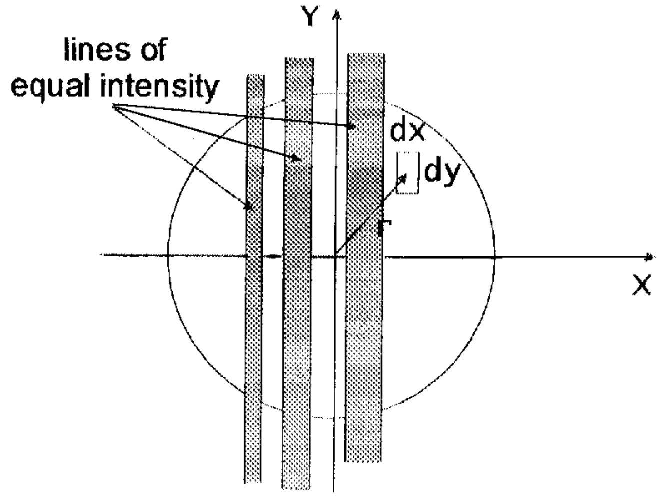

Fig. 2. Summation of the scattered light intensity on the lens area

An infinitely small value of the registered intensity is defined by the relation $\Delta u = vG\Delta S$, where v is the true scattering intensity, G — intensity transmittance coefficient at a given incidence point on the lens, and $\Delta S = \Delta s \Delta t$ — an element of the lens circle area.

In this case, the intensity measured by the CCD-camera is naturally represented by the formula

$$u\,(x) = \iint_S vG\, ds\, dt, \quad x \in [a,b], \tag{4}$$

where x is the coordinate of the lens center.

We make some comments about the functions v and G. In the general case, the true scattering intensity v is a function of two variables s and t. But, in our case, the scattering by an isotropic film of nanoporous TiO_2 is axisymmetric, and v depends only on a polar angle θ. Therefore, at a sufficient distance from the axis $\theta = 0$, where the curvature of equal-intensity conic surfaces is small, ν can be considered as a function of one variable, i.e. $v = v(s)$. We can also naturally assume that the intensity transmittance coefficient at the point of the lens G depends only on the radius $r \in [0, R]$. With regard for these comments, we can write formula (4) in the following form:

$$u\,(x) = \int_{x-R}^{x+R} \int_{-\sqrt{R^2-(x-s)^2}}^{\sqrt{R^2-(x-s)^2}} v\,(s)\, G\,(r)\, ds\, dt. \tag{5}$$

Taking $r \in [0, R]$ as an integration variable, we get

$$u\,(x) = \int_{x-R}^{x+R} v(s)\, ds \int_{|x-s|}^{R} G\,(r)\, \frac{2r}{\sqrt{r^2-(x-s)^2}} dr, \tag{6}$$

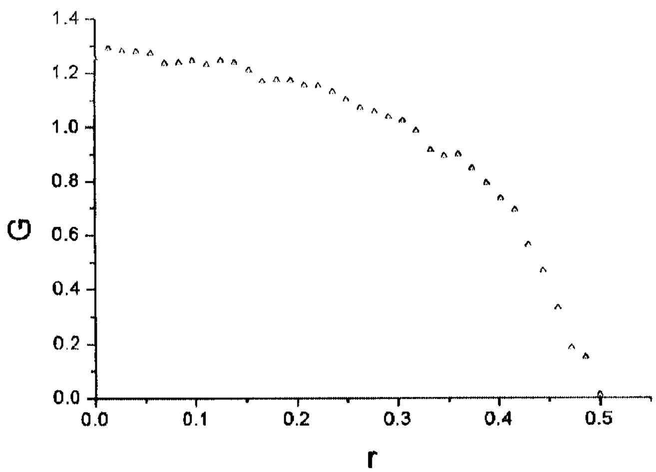

Fig. 3. Lens transmittance measured along its diameter

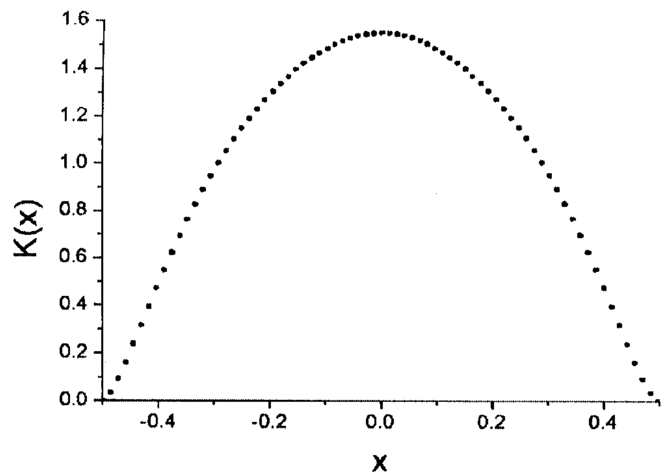

Fig. 4. Apparatus function

or

$$u\,(x) = \int\limits_{-R}^{R} K(s)\,v\,(x-s)\,ds, \qquad (7)$$

where

$$K(s) = \int\limits_{|\,s\,|}^{R} G(r)\frac{2r}{\sqrt{r^2-s^2}}\,dr. \qquad (8)$$

The posed task (7) is known as the problem of reduction to the ideal apparatus. It is important to emphasize that its solution is accompanied by the serious problems concerned with the errors of measurement. As was assumed above, the apparatus function $K(x,s)$ and registered function $u(x)$ which are required to define the sought function $v(s)$ are known exactly. In reality, these two functions can be obtained only via the experimental measurement of $u(x)$ and $G(r)$ by the CCD-camera. These measurements always contain errors concerned with the properties and faults of the measuring tract (defects of optics, inaccurate focusing, etc.).

Expression (8) shows that the function $K(x)$ is a function of experimental values of the intensity transmittance coefficient G. The dependence $G(r)$ was measured using the same experimental setup without a sample, i.e. the laser beam scanned the lens along its diameter. The experimental data on $G(r)$ that are correct to within the normalization factor are given in Fig. 3.

An experimental dependence for $\tilde{K}(x)$ can be obtained from these values with integration according to formula (8). The result is given in Fig. 4.

For the purpose of approximation of the experimental values of the function $\tilde{K}(x)$, we can use the following consideration. Let us assume that $K(x,s) = \delta(x-s)$. Then, according to (3),

$$\int\limits_{a}^{b} \delta(x-s)\,v\,(s)\,ds = v\,(x),$$

i.e. we have the "ideal" measuring tract.

On the other hand, it is known that the Dirac delta-function is the simplest example of a singular generalized function. Moreover, there exist delta-like sequences which converge to $\delta(x)$ in the space of generalized functions K$'(\Omega)$ $(\Omega \subset R^n)$ as $\gamma \to +0$ [4, 5]:

$$\left.\begin{array}{ll} 1. & \frac{1}{\pi}\frac{\gamma}{x^2+\gamma^2} \\ 2. & (\pi x)^{-1}\sin(x/\gamma) \\ 3. & [\gamma/(\pi x^2)]\sin^2(x/\gamma) \\ 4. & e^{-x^2/4\gamma}/(2\sqrt{\pi\gamma}) \end{array}\right\} = K_\gamma(x) \to \delta(x),\ \gamma \to +0. \qquad (9)$$

N	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
r	0	0.014	0.028	0.042	0.056	0.069	0.083	0.097	0.111	0.125	0.139	0.153	0.167	0.181	0.194	0.208	0.222	0.236	0.25
G	1.255	1.29	1.28	1.277	1.272	1.235	1.237	1.242	1.23	1.243	1.238	1.208	1.168	1.174	1.171	1.152	1.15	1.128	1.1

N	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37
r	0.264	0.278	0.292	0.306	0.319	0.333	0.347	0.361	0.375	0.389	0.403	0.417	0.431	0.444	0.458	0.472	0.486	0.5
G	1.068	1.055	1.034	1.02	0.982	0.912	0.888	0.895	0.845	0.789	0.73	0.687	0.561	0.463	0.331	0.184	0.15	0

Here, we imply a weak limit of the sequence of functions $K_\gamma(x)$, $\gamma \to +0$. That is, for an arbitrary function $\varphi(x)$ from the space K of basic functions, the following limit relationship is valid:

$$\lim_{\gamma\to+0} \int K_\gamma(x)\,\varphi(x)\,dx = (\delta,\varphi) = \varphi(0) \tag{10}$$

(the space of basic functions $\mathrm{K}(\Omega)$ consists of all finite functions φ which have continuous derivatives of all orders).

It can be shown that a “hat” function $\omega_\gamma(x)$ [5]

$$\omega_\gamma(x) = \begin{cases} C_\gamma e^{-\gamma^2/(\gamma^2-|x|^2)}, & |x|<\gamma, \\ 0, & |x| \ge \gamma, \quad x\in R^n; \end{cases} \tag{11}$$

where a constant C_γ is chosen to be such that $\int \omega_\gamma(x)\,dx = 1$, i.e.

$$C_\gamma = \left(\gamma^n \int\limits_{|\xi|<1} e^{-\frac{1}{1-|\xi|^2}}\,d\xi\right)^{-1},$$

also forms a delta-like sequence:

$$\omega_\gamma(x) \to \delta(x), \quad \gamma\to+0. \tag{12}$$

According to the definition of convergence in $\mathrm{K}'(\Omega)$, relation (12) is equivalent to the equality

$$\lim_{\gamma\to+0}\int \omega_\gamma(x)\,\varphi(x)\,dx = \varphi(0), \quad \varphi\in \mathrm{K}$$

which follows from the estimation

$$\left|\int \omega_\gamma(x)\,\varphi(x)\,dx - \varphi(0)\right| \le$$

$$\le \int \omega_\gamma |\,\varphi(x)-\varphi(0)\,|\,dx \le$$

$$\le \max_{|x|\le\gamma} |\,\varphi(x)-\varphi(0)\,| \int \omega_\gamma(x)\,dx =$$

$$= \max_{|x|\le\gamma} |\,\varphi(x)-\varphi(0)\,|$$

and from the continuity of the function $\varphi(x)$.

The set of apparatus functions well-known in physics, which includes the diffractional

$$K(x) = \frac{1}{\gamma}\left[\frac{\sin(\pi x/\gamma)}{\pi x/\gamma}\right]^2 \tag{13}$$

(the half width of an apparatus curve is defined by the relation $a = 0.886\gamma$), slit-like

$$K(x) = \begin{cases} 1/a & \text{when} \quad |x|/a \le 1/2 \\ 0 & \text{when} \quad |x|/a > 1/2 \end{cases} \tag{14}$$

(a — the width of a slit image), Gaussian

$$K(x) = \frac{2}{a}\sqrt{\frac{\ln 2}{\pi}}\exp\left\{-4\ln 2\frac{x^2}{a^2}\right\}, \tag{15}$$

dispersive

$$K(x) = \frac{a/2\pi}{x^2+(a/2)^2}, \tag{16}$$

Dirichlet

$$K(x) = \frac{\sin(x/a)}{\pi x}, \tag{17}$$

exponential

$$K(x) = \frac{\ln 2}{a}\exp\left\{-2\frac{|x|}{a}\ln 2\right\}, \tag{18}$$

triangle

$$K(x) = \begin{cases} a^{-1}(1-|x|/a) & \text{when} \quad |x|/a \le 1 \\ 0 & \text{when} \quad |x|/a > 1 \end{cases} \tag{19}$$

functions, forms a subset of delta-like sequences (pulsed functions). Vice versa, the pulsed functions, in particular, a “hat” function

$$K_\gamma(x) = \begin{cases} C_\gamma e^{-\gamma^2/(\gamma^2-x^2)}, & |x|<\gamma, \\ 0, & |x|\ge\gamma, \quad x\in(-\infty,\infty); \end{cases} \tag{20}$$

the Stieltjes function

$$K_\gamma(x) = \frac{2}{\pi\gamma}\,\frac{1}{e^{x/\gamma}+e^{-x/\gamma}} = \frac{1}{\pi\gamma}\mathrm{ch}^{-1}\frac{x}{\gamma}, \tag{21}$$

and the function

$$K_\gamma(x) = \frac{2}{\pi}\frac{\gamma^3}{(x^2+\gamma^2)^2}, \tag{22}$$

can be used as apparatus functions.

From the point of view of practical applications, the “hat” function (20) is of particular interest because it has the following important properties [6]. Firstly, it is compact by definition, i.e. $K_\gamma(x) = 0$, $|x| \ge \gamma$. Second, it is infinitely differentiable with respect to x. Indeed, it is obvious for $|x| < \gamma$ and for $|x| > \gamma$ (in the last case, all derivatives are equal to zero). Therefore, it suffices to verify that its derivatives of any order for $x < \gamma$ go to zero as $x \to \gamma$.

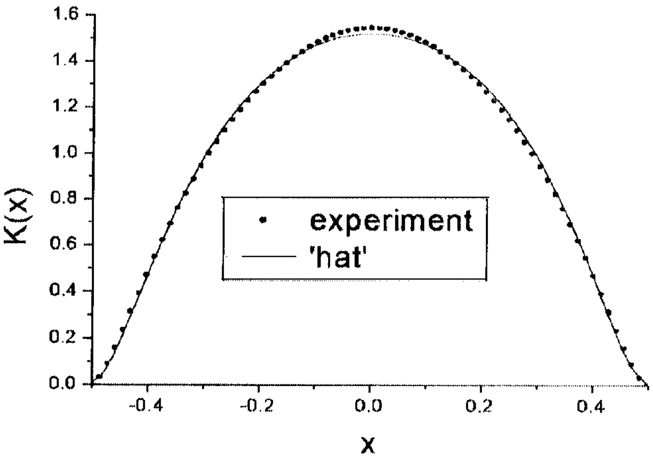

Fig. 5. Apparatus function and its approximation with the "hat" function

The function $K_\gamma(x)$ (20) is continuous at $x = \gamma$ because

$$K_\gamma(\gamma + 0) = 0 = K_\gamma(\gamma)$$

and

$$K_\gamma(\gamma - 0) = \lim_{x\to\gamma-0} C_\gamma e^{-\gamma^2/(\gamma^2-x^2)} = 0,$$

since $\lim\limits_{x\to\gamma-0}\left(-\frac{\gamma^2}{\gamma^2-x^2}\right) = -\infty.$

A derivative of the function $K_\gamma(\gamma)$ exists and is equal to zero:

$$\lim_{x\to\gamma+0}\frac{K_\gamma(x)-K_\gamma(\gamma)}{x-\gamma} = \lim_{x\to\gamma+0} 0 = 0,$$

$$\lim_{x\to\gamma-0}\frac{K_\gamma(x)-K_\gamma(\gamma)}{x-\gamma} = \lim_{x\to\gamma-0}\frac{C_\gamma e^{-\gamma^2/(\gamma^2-x^2)}}{x-\gamma} =$$

$$= C_\gamma \lim_{x\to\gamma-0}\frac{(x-\gamma)^{-1}}{e^{\gamma^2/(\gamma^2-x^2)}} =$$

$$= C_\gamma \lim_{x\to\gamma-0}\frac{-(x-\gamma)^{-2}}{\left(\frac{2x\gamma^2}{(\gamma-x)^2(\gamma+x)^2}e^{\gamma^2/(\gamma^2-x^2)}\right)} =$$

$$= C_\gamma \lim_{x\to\gamma-0}\left(-\frac{(\gamma+x)^2}{2x\gamma^2}e^{-\gamma^2/(\gamma^2-x^2)}\right) = 0.$$

Thus, a limit exists and it is equal to zero

$$\lim_{x\to\gamma}\frac{K_\gamma(x)-K_\gamma(\gamma)}{x-\gamma} = K'_\gamma(\gamma).$$

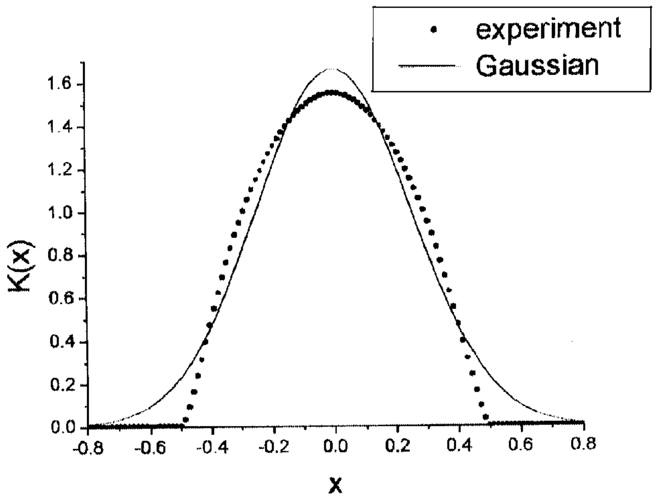

Fig. 6. Apparatus function and its approximation with a Gaussian function

The following relation is valid:

$$\lim_{x\to\gamma-0} K'_\gamma(x) = \lim_{x\to\gamma-0} C_\gamma \frac{2x\gamma^2}{(\gamma^2-x^2)^2}e^{-\gamma^2/(\gamma^2-x^2)} = 0.$$

Thus, the first derivative $K'_\gamma(x)$ of the function (20) exists and is continuous at an arbitrary x. The existence and continuity of the following derivatives are verified in a similar way.

The compactness and infinite differentiability of the apparatus function $K_\gamma(x)$ distinguish it, in principle, from the apparatus functions (13-19). For example, the slit-like apparatus function (14) and triangle one (19) are compact, but they do not have continuous first derivatives. On the other hand, the Gaussian (15) and diffractional (13) apparatus functions are infinitely differentiable but not compact.

The fitting of the experimental values of $\tilde{K}(x)$ with the "hat" function $K_\gamma(x)$ (20) resulted in the parameters:

$$C_\gamma = 4.13, \quad \gamma = 0.545.$$

The best approximation of the values of $\tilde{K}(x)$ with the function $K_\gamma(x)$ in the metric $L_2(-0.5, +0.5)$ is shown in Fig. 5.

For comparison, the best approximation of $\tilde{K}(x)$ with the Gaussian function is shown in Fig. 6.

It is evident that the "hat" function $K_\gamma(x)$ is much more appropriate for the approximation of the apparatus function values.

Returning to the problem of reduction, let us consider the existing approaches to solving the integral equation (7) (a convolution-like equation) when the function $u(x)$ and apparatus function $K(x)$ are known.

We should emphasize that the peculiarity of the considered inverse problems (3), (7) consists in the integral nature of the observed function $u(x)$ and in its insensibility to significant changes of the function $v(x)$ if these changes compensate one another. This means that two essentially different curves of $v(x)$ may correspond to close observed functions. Moreover, as long as the registered functions are always measured with some error, we meet the problem to find an approximate solution of the inverse problem (7) which would be close to the true solution. This is exactly the main problem for the mathematical interpretation of experimental results.

It was shown in [7] that the solution of Eq. (7) exists and is unique in the case where the Fourier transform of an apparatus function doesn't turn to zero on any finite interval.

The classical method of solution of convolution-like equations, which uses the direct and inverse Fourier transformations, consists in the following [8].

Let $u(x) \in L_2(-\infty,\infty)$, $K(x) \in L_1(-\infty,\infty)$, $v(x) \in L_1(-\infty,\infty)$, then the solution of Eq. (7) is defined by

$$v(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{U(\omega)}{\mathrm{K}(\omega)} \exp(-ix\omega)\,d\omega, \quad x \in (-\infty\,,\infty), \quad (23)$$

where $U(\omega)$ and $\mathrm{K}(\omega)$ are the Fourier transforms of the observed $u(x)$ and apparatus $K(x)$ functions, i.e.

$$U(\omega) = \int_{-\infty}^{\infty} u(x)\exp(i\omega x)\,dx,$$

$$\mathrm{K}(\omega) = \int_{-\infty}^{\infty} K(x)\exp(i\omega x)\,dx.$$

If the Fourier transforms $U(\omega)$, $\mathrm{K}(\omega)$ tend consistently to zero as $\omega \to \infty$ in such way that

$$\lim_{\omega\to\infty} \frac{U(\omega)}{\mathrm{K}(\omega)} = 0,$$

and integral (23) converges, then the solution $v(x)$ exists. It is unique and defined by formula (23). That is, for precise $u(x)$ and $K(x)$, the first two points of the problem correctness definition by Hadamard can be satisfied.

But, even at this stage, there are difficulties to use formula (23), since the manipulation with infinite limits is possible only if we have analytical expressions for $U(\omega)$ and $\mathrm{K}(\omega)$. There is no analytic representation for the Fourier transform of a "hat" function $K_\gamma(x)$.

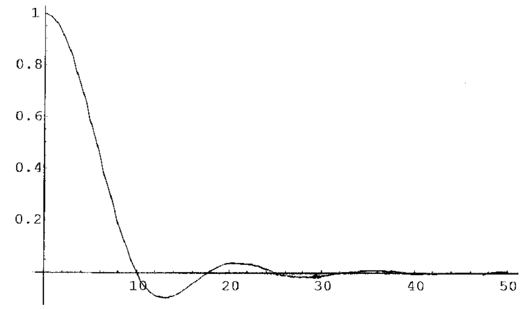

Fig. 7. Cosine Fourier spectrum of the "hat" function $K_{0.5}(x)$

It has already been mentioned that serious problems arise due to the errors of measurement of the scattered light distribution, which originate from the properties and faults of the measuring tract as well as from the quantum nature of the registered light energy. In the presence of errors of measurement, the resulting curve can be represented as $\tilde{u}(x) = u(x) + \zeta(x)$, where $\zeta(x)$ is a noise. It contains, in particular, a white noise component which makes $\widetilde{U}(\omega)$ tending not to zero when $\omega \to \infty$ but to some constant that depends on the level of white noise. Then $\lim_{\omega\to\infty} \frac{\widetilde{U}(\omega)}{\mathrm{K}(\omega)} = \infty$ and, according to (23), no solution of Eq. (4) exists. Thus, high harmonics in the solution $v(x)$ are sensitive even to small errors in $u(x)$. This yields that ignoring the instability of the problem of reduction to the ideal apparatus may lead to a wrong interpretation of experimental results.

As was mentioned above, there is no analytic expression for the Fourier transform of the "hat" function $K_\gamma(x)$. But a numerical calculation of the Fourier transform of this function doesn't meet any difficulties. Fig. 7 shows the results of numerical calculations for the spectrum of $K_{0.5}(x)$.

Its first several zeroes have the values:

$$w_1 = 9.993, \quad w_2 = 17.777, \quad w_3 = 25.198.$$

In conclusion the main results of the work can be formulated in the following way:

— a mathematical model of an experimental setup for light scattering measurement in the form of an integral 1st kind Fredholm equation has been obtained;

— a kernel of the integral equation (apparatus function) $\widetilde{K}(x)$ has been defined in a rather simple way via the experimental values of the transmittance coefficient $G(r)$ of the lens measured along its diameter;

— experimental values of the apparatus function $\widetilde{K}(x)$ have been perfectly fitted by the 'hat' function $K_\gamma(x)$.

A relative error of the approximation is $||\delta K||_{L_2} = 0.01$;
— zeroes countability of the Fourier image of apparatus function allows the application of the regularization method for the analysis of a mathematical model of the experimental setup.

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1. *Starkov V.N.* // Constructive Methods of Computational Physics in Interpretation Problems. — Kyiv: Naukova Dumka, 2002 (in Russian).
2. *Tikhonov A.N., Arsenin V.Ya.*// Methods of Solution of Ill-Posed Problems. — Moscow: Nauka, 1979 (in Russian).
3. *Physics* Encyclopedia/Ed. by A.M.Prokhorov.— Moscow: Soviet Encyclopedia, 1984 (in Russian).
4. *Sadovnichy V.A.*// Theory of Operators. — Moscow: Moscow Univ. Press, 1986 (in Russian).
5. *Vladimirov V.S.*// Equations of Mathematical Physics. — Moscow: Nauka, 1988 (in Russian).
6. *Vladimirov V.S.*// Generalized Functions in Mathematical Physics. — Moscow: Nauka, 1979 (in Russian).
7. *Khalfin L.A.* // Opt. Spectr. — 1969. — 26. — P. 1065—1067.
8. *Gakhov F.D., Chersky Yu.I.*// Convolution-like Equations. — Moskow: Nauka, 1978 (in Russian).

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МАТЕМАТИЧНЕ МОДЕЛЮВАННЯ ЕКСПЕРИМЕНТАЛЬНОЇ УСТАНОВКИ ДЛЯ ВИМІРЮВАННЯ РОЗСІЯННЯ СВІТЛА ПОРИСТИМИ ПЛІВКАМИ

А.О.Борщ, М.С.Бродин, В.Я.Гайворонський, П.В.Максютенко, В.М.Старков

Р е з ю м е

Побудована математична модель експериментальної установки для вимірювання розсіяння світла в матеріальному середовищі. Наявність такої моделі дозволяє за експериментальними даними відновити інтенсивність розсіяння, не спотворену приладом. Обгрунтовано використання як апаратної апроксимуючої функції “шапочка”.