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Comparison of the results of refractometric measurements in the process of diffusion, obtained by means of the background-oriented schlieren method and the holographic interferometry method

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Abstract. **The results of the study of interdiffusion of two liquids, obtained using the holographic recording scheme with a nonstationary reference wave with the frequency linearly varying in space and time are compared with the results of correlation processing of digital photographs, made with a random background screen. The spatio-temporal behaviour of the signal in four basic representations ('space–temporal frequency', 'space–time', 'spatial frequency–temporal frequency' and 'spatial frequency–time') is found in the holographic experiment and calculated (in the appropriate coordinates) based on the background-oriented schlieren method. Practical coincidence of the results of the correlation analysis and the holographic double-exposure interferometry is demonstrated.**

Keywords: *holographic interferometry, nonstationary reference wave, background-oriented schlieren, digital image correlation, diffusion of liquids.*

1. To visualise optical inhomogeneities and measure their value the background-oriented schlieren (BOS) method is widely used. In Ref. [1] this method was applied to solve refractometric problems using a flow above a flame as an optical nonuniformity, in Refs [2, 3] it was used for measuring distortion and chromatic aberration in optical systems, and in Ref.[4] – for determining deformations of transparent bodies. The precision of determining the relative shifts of image pairs in the matrix of a photographic camera can be as small as hundredths of a pixel. Actually, it is a method of measuring the optical density gradient. It is interesting to compare the results of studying a certain process, giving rise to medium optical inhomogeneity, by means of the correlation method with the results obtained using other methods.

2. In the present paper we compare the results of studying the interdiffusion of two colourless liquids. The data were obtained by correlation processing of digital photographs of a random background screen, made in white light through the cell where the diffusion process occurred. As a standard for comparison we have chosen the results for the laser radiation passing through a cell with interdiffusion, obtained by us much earlier using the method of double-exposure holographic interferometry with a nonstationary reference wave [5–8]. The paired liquids significantly differed in density; therefore, in the cell their separation occurred rather quickly, giving rise to an interface layer, perpendicular to the gravity force direction, so that the system acquired quasi-one-dimensional character. This fact allowed simplification of the experimental scheme. In principle, the holographic method can be applied for objects with complex spatial behaviour, cutting the region of interest with a slit diaphragm, but the correlation method allows the study of such objects without such operations, directly analysing two-dimensional images. In the present work we performed the comparison within the frameworks of the process, considered in the old holographic experiment, i.e., the quasi-one-dimensional case was considered.

3. Under the conditions of interdiffusion of two transparent liquids with different refractive indices, the refractive index $n(t,x)$ of the medium changes, making it to be a function of time and spatial coordinate. Let us assume that the refractive index depends only on the concentration N of one substance, i.e.,

$$n = \beta N, \tag{1}$$

and the behaviour of the concentration is described by the solution of the diffusion equation with the diffusion coefficient independent of the concentration (this pair of liquids will be referred to as ideal)

$$\frac{1}{D}\frac{\partial N}{\partial t} = \frac{\partial^2 N}{\partial x^2}. \tag{2}$$

The function

$$\varphi(t,x) = dn(t,x), \tag{3}$$

where d is the cell thickness, has the dimension of length and is the studied part of the optical path of the light beam to the recording system, varying in the process of diffusion, both in the case of holographic information recording and in the case of correlation measurements. The cell is assumed to be thin, so that one can neglect the transverse shift of the beam in the process of its propagation. Obviously, under these conditions the refractive index will be described by the same diffusion equation (2) with the same diffusion coefficient. In Ref. [5] we used the notion of an instantaneous temporal frequency at a given spatial point (see also [9])

$$\nu = -\frac{1}{\lambda}\frac{\partial \varphi}{\partial t} = -\frac{d}{\lambda}\frac{\partial n}{\partial t}, \tag{4}$$

and, similarly, the local spatial frequency, or angular coordinate (related to the gradient of the refractive index)

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$$q_x = \frac{\partial \varphi}{\partial x} = d\frac{\partial n}{\partial x}, \tag{5}$$

which under the given geometry of experiment is proportional to the refractive index gradient, since the variations occur only along one spatial coordinate perpendicular to the interface between the liquids, the layer thickness remaining constant.

4. In the holographic setup the signal recording was implemented in accordance with the Leith scheme using a nonstationary reference wave with the frequency, linearly dependent on the coordinate and time [10]. The frequency varied along the horizontal coordinate (along the interface between the liquids). Such a wave can be presented as a wave with a planar wave front that rotates around the axis, parallel to the plane of the hologram, separated from it by a certain distance. The recording was executed according to the double-exposure interferometry scheme, i.e., the recording of the signal wave (the plane wave passed through the studied cell) was superposed with the similar record of a signal in the absence of the cell, made at the other time (the additional signal was recorded). After two expositions, the hologram was photochemically processed. The information about the spatial and temporal distributions of the signal was recorded along the vertical and the horizontal coordinate of the hologram, respectively. The complete information about the field was thus recorded in the hologram. After reconstructing the recorded field with a stationary reference wave all this information was contained in the reconstructed wave. In Refs [5–8] the analysis of the field structure recorded in the hologram was carried out and it was shown that this field can be reconstructed in different representations. The spatial and temporal field distributions are independent, and using astigmatic optics one can obtain their four 'canonical' forms, in which for every coordinate either the appropriate coordinate distribution, or its Fourier transform is presented. These distributions contain complete information about the spatio-temporal behaviour of the signal in all the following four representations: 'space–temporal frequency', 'space–time', 'spatial frequency–temporal frequency' and 'spatial frequency–time'. In each of the representations the specific features of the diffusion process manifest themselves.

For correlation processing, the cell with diffusing liquids was photographed against the background of a random screen. The experiments were carried out with aqueous solutions of KCl and ethyl alcohol in different concentrations. The aim of the present paper is to show the possibility of using the correlation method for obtaining the abovementioned four basic representations of the signal and determining the diffusion coefficient.

5. As in Refs [6–8], no special measures were necessary to facilitate thorough layering of one liquid over another and prevent their mixing. Under the action of gravity, the liquids quickly and spontaneously separated hydrodynamically due to the density difference, forming a relatively narrow interface layer. Then the diffusion from the stabilised initial state was observed.

The background screen, which was photographed through the cell with the diffusion process, was a square regular grid with random black and white filling of the cells (the filling probability being 0.5). The scale of photography was chosen such that the image size of one cell at the photographic camera matrix was 2.5 pixels. This size is optimal from the point of view of the signal-to-noise ratio [11]. The diffusion process occurred in the cell with the height 4 cm and the thickness 1 mm. The scheme of photographic recording is presented in Fig.1. The separation between the background pattern and the cell was l = 37 cm, and between the background pattern and the photographic camera L = 51 cm. The camera used was Olympus μ Tough, with the focal length f = 7 mm; the maximal displacement of the image elements in the presented cases amounted to 5–11 pixels.

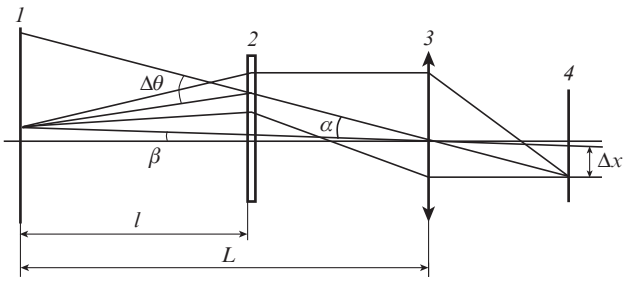

Figure 1. Recording scheme: (*1*) background pattern; (*2*) cell with the studied liquids; (*3*) camera objective; (*4*) receiving matrix plane.

6. The beam deflection $\Delta\theta$, caused by its transmission through the cell, leads to the shift of the corresponding region of the random pattern image by Δx in the plane of the camera matrix. Assuming the angles α and β to be small, for the known shift Δx one can calculate the angle

$$\Delta\theta \approx \Delta x \frac{L-f}{lf} \tag{6}$$

and the refractive index gradient of the planar cell content

$$\frac{\mathrm{d}n}{\mathrm{d}x} \approx \Delta x \frac{L-f}{Lfd}. \tag{7}$$

The role of the reference image was played by the photograph of the screen through the cell filled with the liquid of one sort. Then with some time intervals the photographing was repeatedly carried out through the cell with diffusing liquids. The dependence of Δx on the vertical coordinate was determined by correlation scanning of the pairs of photographs (the one made at a definite moment of time and the reference one) with a 256 × 32 pixel window stretched in the horizontal direction. This shape of the window provided the locality of measurements in the vertical direction, in which the refractive index was varying, and their sufficient statistical stability (due to the large horizontal size). The results of measuring the refractive index gradient, proportional to the local spatial frequency or angular coordinate (using the terminology of Refs [6–8]) are shown in Fig. 2. The abscissa axis is directed downwards along the centre of the cell.

After the integration, the profile of the refractive index is reconstructed (Fig. 3). The difference between the refractive index at the given depth in the cell and the refractive index of the liquid with smaller optical density at the origin of the coordinate frame is calculated. The refractive index of distilled water is $n_1 \sim 1.333$, which yields $n_2 \sim 1.364$ for the 3 M solution of KCl (the tabulated value is 1.362 [12]).

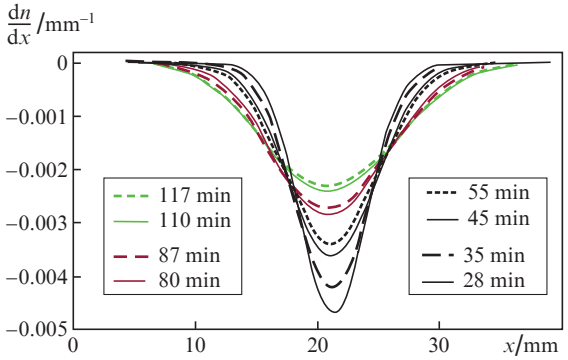

Figure 2. Dependences of the refractive index gradient on the coordinate for the pair 'water–3M solution of KCl' at different times after the beginning of diffusion.

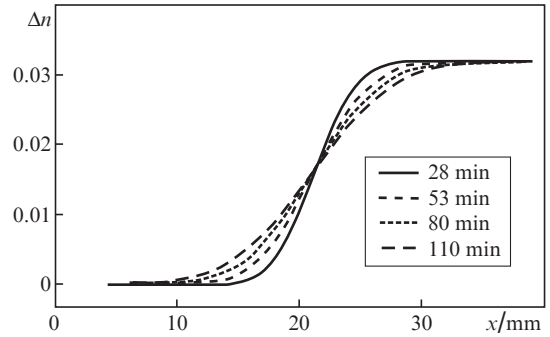

Figure 3. Dependences of the refractive index difference on the coordinate for the pair 'water–3 M solution of KCl' at different times after the beginning of diffusion.

7. One can calculate the temporal behaviour of the refractive index time derivative at different spatial points and at different moments of time. The theoretical dependence of this quantity on the coordinate from [6–8] is presented in Fig. 4a. Figure 4b (the photo of the image reconstructed from the double-exposured hologram) shows the field distributions of the first type in the coordinates 'temporal frequency–space',

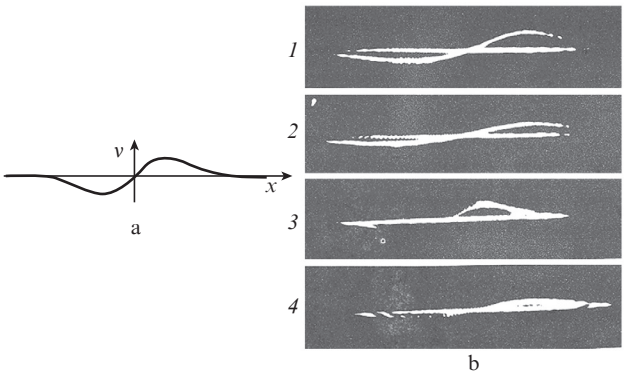

Figure 4. Field distribution in the coordinates 'space–temporal frequency' in the holographic experiment: (a) theoretical dependence and (b) experimental data for the solutions of H_2SO_4 [the time after the beginning of diffusion process (*1*) 19 min 45 s and (*2*) 34 min] and KCl [(*3*) 5 min 20 s and (*4*) 20 min 10 s]. Horizontal lines plot the additional signal having a zero frequency.

experimentally determined in Refs [6–8] for the solutions H_2SO_4 (3.71 M) and KCl (3 M). The abscissa of each point represents the spatial coordinate, and the ordinate represents the instantaneous frequency. The brightness of point in the photo corresponds to the brightness of light.

In correlation processing of digital photographs the 'space–temporal frequency' distribution corresponds to the dependence of the refractive index time derivative on the coordinate, resulting from processing the pairs of photographs made with some temporal interval. The dependence of the refractive index variation rate on the coordinate, calculated from the digital photographs, is shown in Fig. 5 for some moments of time. The qualitative agreement with the data of holographic experiment is seen.

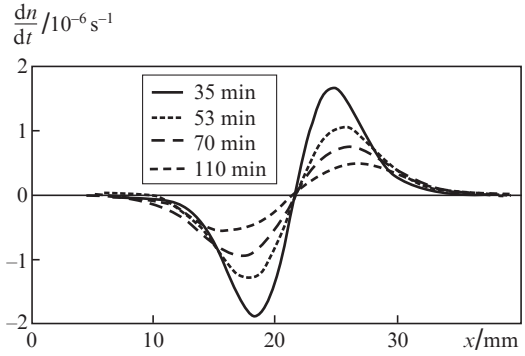

Figure 5. Dependences of the time derivative of the refractive index on the spatial coordinate at given times after the beginning of diffusion.

8. The distribution of the second kind in the 'space–time' coordinates after the reconstruction of the holographic record had the form of the interference of the signal, modulated by the cell, and the unmodulated signal from another exposure. In the pattern one can clearly see interference fringes, 'drawing' the spatio-temporal behaviour of the refractive index [6–8] (Fig. 6a). Actually, it can be said that each interferogram is a few tens of continuous time-resolved frames. At every moment of time the interference pattern is recorded, from which one can reconstruct the depth (vertical) distribution of phase $\Phi = (2\pi/\lambda)\varphi(x)$, thus making it possible to determine the refractive index profile. Comparing two distributions close enough in time, one can determine the time derivative of the refractive index, characterised by the fringe slope at the considered point. These interference patterns were further used to calculate other representations of the signal [6–8]. Figure 6 b shows model interferograms, which were calculated from the refractive index profile obtained by the correlation method using the digital images taken at different times. The abscissa axis is related to time, and the ordinate axis – to the spatial coordinate. The wavelength for the model calculation was taken equal to 1500 nm. In constructing each of the model interferograms we used phase profiles obtained from two images taken at certain time intervals. These intervals are assigned to the boundaries of the relevant interferograms. In the interval, the phase profile was obtained by linear interpolation of the boundary profiles (fringe curving was considered negligible).

9. The third type of the field representation reconstructed from the hologram corresponds to a two-dimensional image, connecting time and spatial frequencies (Figs 7a–7c). Each

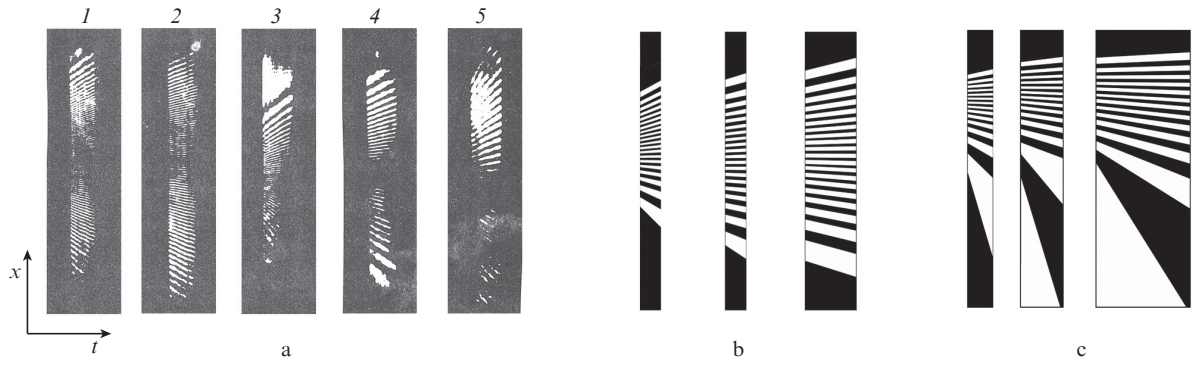

Figure 6. Field distributions in the coordinates 'space–time': data of the holographic experiment for the solutions of H_2SO_4 [the time after the beginning of the diffusion process (*1*) 19 min 45 s and (*2*) 34 min] and KCl [(*3*) 5 min 20 s, (*4*) 20 min 10 s and (*5*) 60 min] (a) and model interferograms, calculated from the refractive index profile in the correlation experiment for (b) KCl and (c) ethyl alcohol.

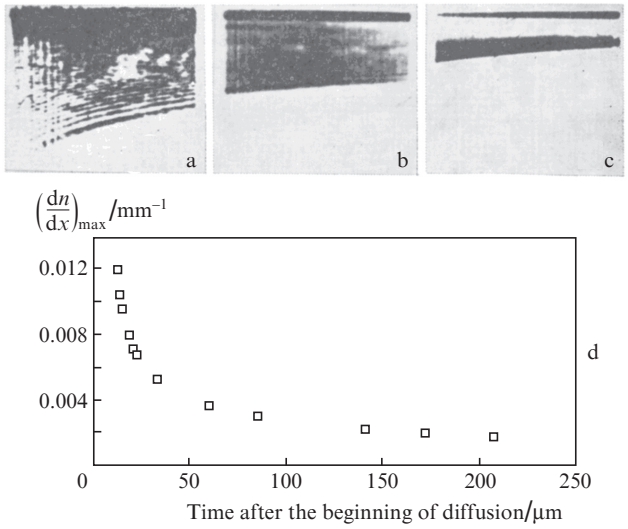

Figure 7. Field distributions in the coordinates 'spatial frequency (ordinate axis)–time (abscissa axis)' in the holographic experiment for the solution of KCl for the exposure time (a) 5 min 20 s–17 min 55 s, (b) 20 min 10 s–32 min 45 s and (c) 60 min–160 min 44 s; (d) is the time dependence of the maximal gradient of the refractive index in the process of diffusion for the solution of ethyl alcohol (correlation experiment).

point of the images contains information about the field of the wave running at a given time in a given direction. The top horizontal line is the line of zero frequencies, reconstructed from the hologram that was obtained during the second exposure. The bands decreasing in width is a pattern of the spectrum of spatial frequencies for the diffusion region. Interference fringes are clearly visible in Fig. 7 and are associated with the interference of the parts of the wavefront going in the same direction from two different areas of the cell [6–8].

In the case of the BOS method this picture corresponds to the time dependence of the refractive index gradient. Figure 7d presents the dependence of the maximal gradient of the refractive index. This dependence has been used for optical determination of the diffusion coefficient [13].

10. Finally, it is possible to obtain the fourth type of signal representation in the coordinates 'spatial frequency–temporal frequency'. In the correlation experiment it corresponds to the representation in the coordinates 'refractive index gradient–refractive index time derivative'. For the diffusion process this dependence has the form of a parametric curve for a certain moment of time. The dependence, obtained from the diffusion equation, is shown in Fig. 8a. In the holographic recording scheme the field distribution pattern in the coordinates 'spatial frequency–temporal frequency' was obtained both optically (in the focal plane of the camera, focused at infinity, Fig. 8b) and numerically by processing the interferograms (Fig. 8c). The corresponding dependences, obtained as a result of correlation processing of digital photographs of the diffusion process, are presented in Fig. 8d. Noticeable asymmetry of the loops in Figs 8b, d is most probably due to the variability of the diffusion coefficient under the variation of the concentration of the solution in the transient region. It is more expressed when use is made of sulphuric acid (Fig. 8b), for which the dependence of the diffusion coefficient on the concentration is stronger. This asymmetry is indicated by the slope of the tangent to the curve at zero temporal frequency and the maximal angular coordinate. In this case equality (2) is not valid in the region of a maximal gradient at a zero spatial frequency.

The algorithm of obtaining the dependence shown in Fig. 8c is thoroughly described in Ref. [6]. It is based on using a microscope to measure the coordinates of the maximal blackening positions of the interference fringes in some vertical section of the distributions, shown in Fig. 6a. The time derivatives were determined by the coordinates of one fringe in two sections. The refractive index profile at a certain moment of time was found using the polynomial interpolation by means of the least squares method in the array of fringe coordinates for the corresponding section. In this case the minimised functional incorporated the derivatives of a few lower orders of the approximating polynomial with selected weights to stabilise the values of the first and the second spatial derivative. Then the spatial derivative was calculated using the obtained polynomial. In the same way the approximating polynomial for the profile of the time derivative of the refractive index was determined. The pair of numbers obtained in this way (the time and coordinate derivatives at one spatial point) is the pair of coordinates of the appropriate point in Fig. 8c.

11. To obtain the diffusion coefficient it is most convenient to present the data of measurements in the form of dependence of the refractive index time derivative on its second spatial derivative. If the refractive index behaves in correspondence with the diffusion equation (2), this should be

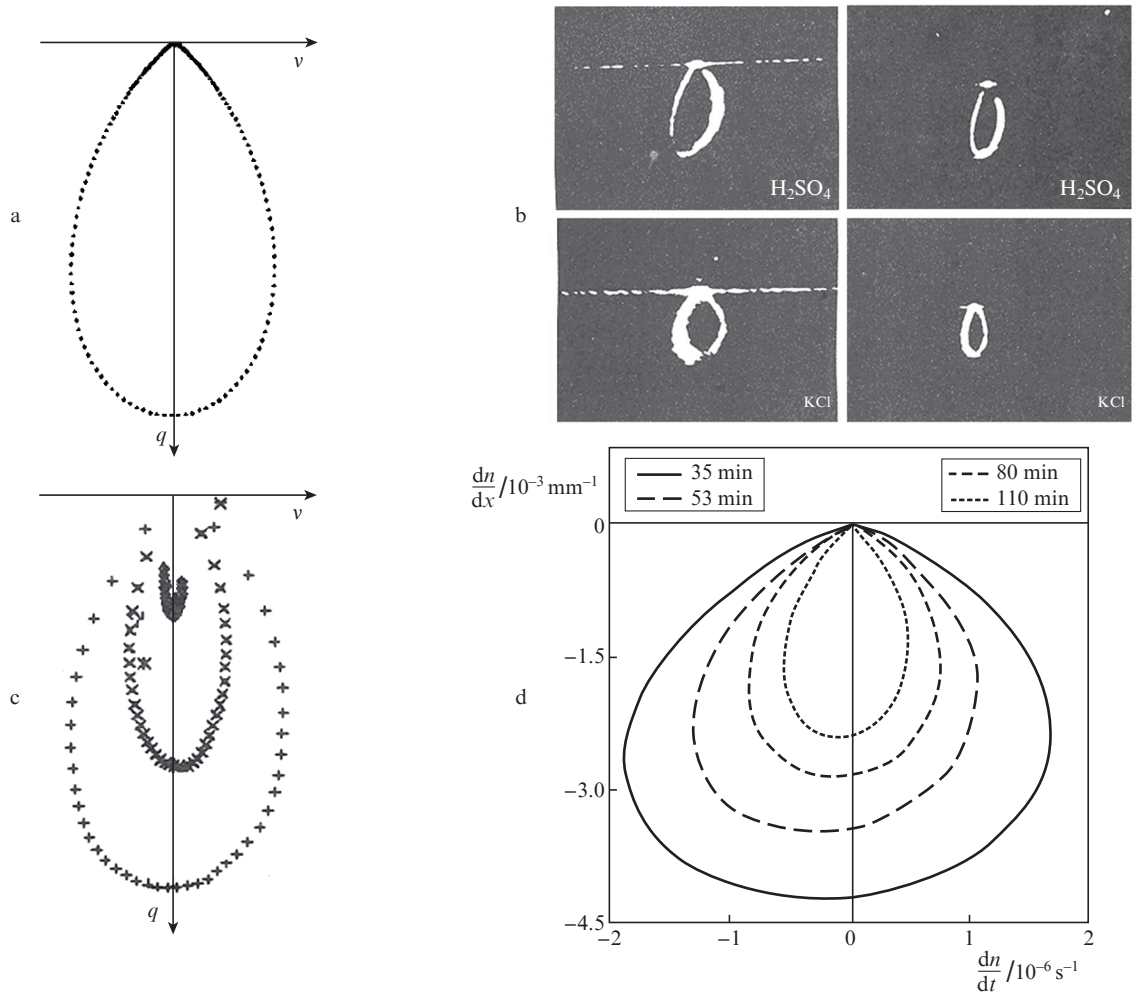

Figure 8. Field distributions in the coordinates 'spatial frequency (ordinate axis)–temporal frequency (abscissa axis)' in the holographic experiment: (a) theoretical dependence and (b) experimental data for the solutions H_2SO_4 and KCl, as well as (c) characteristics for the KCl solution, calculated from the interferograms; (d) is the dependence of the refractive index gradient on the time derivative of the refractive index, calculated from the data of the BOS method.

the proportionality dependence. In this case, the initial profile of the solution concentration is absolutely unimportant. In Fig. 9 for the KCl solution the dependences of the first time derivative of the refractive index on the second coordinate derivative, obtained in the experiment with holographic recording of the diffusion process and by processing the data of BOS method are presented. In the case of the holographic experiment the second derivative was calculated using the interpolation polynomial described above. In all Figures the dependences sufficiently well correspond to the proportional ones. For the holographic experiment at a short time passed from the beginning of the process (Fig. 9a) at the edges of the dependences the loop-like behaviour takes place, i.e., the points of forward and backward traces do not coincide. This is due to the use of the interpolating polynomial for calculating derivatives with small number of experimental points (30–40). In the correlation experiment the number of points is essentially larger, which, probably, explains the absence of the effect manifestations.

The diffusion coefficient of the KCl solution, obtained for the considered moments of time in the holographic experiment, amounts to 1.7×10^{-3} mm^2 s^{-1} for the dependence in Fig. 9a and 1.8×10^{-3} mm^2 s^{-1} for the dependence in Fig. 9b, which corresponds to the tabulated values at the temperature 18.5 °C [12]. According to the data of the experiment, carried out using the BOS method (Figs 9c, 9d) the diffusion coefficient for KCl solution is equal to 1.99×10^{-3} mm^2 s^{-1}, which corresponds to a higher temperature (20–22 °C). The issue of some disagreement between the two branches of the dependence requires special consideration. As in Refs [6–8], for the KCl solution both derivatives ($\partial n/\partial t$ and $\partial^2 n/\partial x^2$) are close to zero in the vicinity of the interface, which indicates weak dependence of the diffusion coefficient on the concentration. Note that the initial distribution of the refractive index is absolutely unimportant, because it is not needed to solve the diffusion equation, since we use the numerical values of the left-hand and the right-hand side of the diffusion equation ($\partial n/\partial t$ and $\partial^2 n/\partial x^2$), extracted from the experimental data.

12. From the general considerations it can seem that the method of double-exposure holographic interferometry yields the correlation method. However, it should be kept in mind that the former was based on the old methods of acquiring data for subsequent processing. On the contrary, the correlation method was completely based on the modern measuring techniques of image recording followed by the computer processing. Nevertheless, the results obtained by us allow the conclusion that the correlation method is not inferior to the holographic one in the precision of reconstructing the object

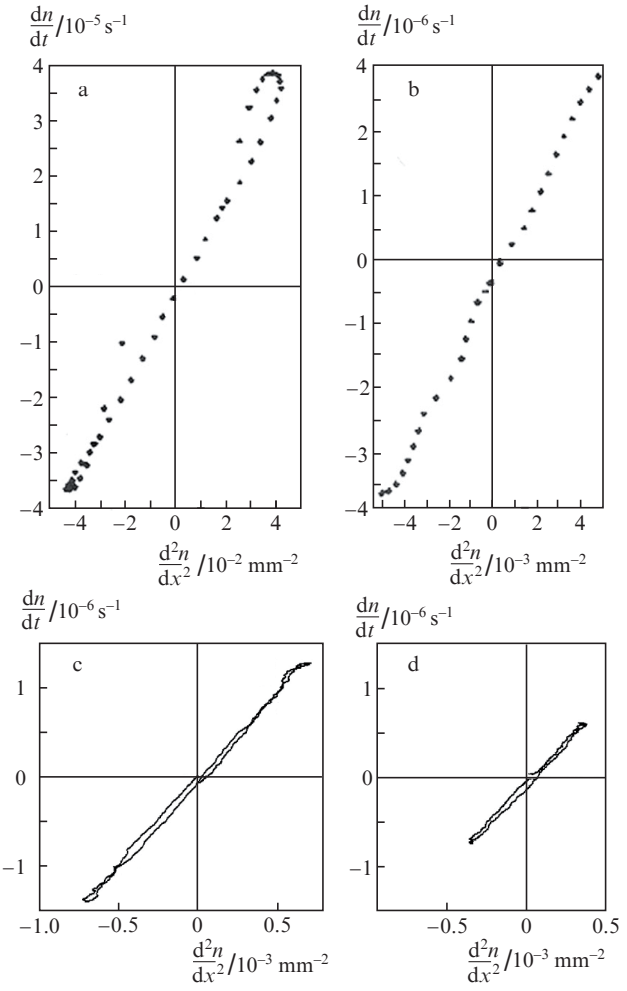

Figure 9. Dependences of the time derivative of the refractive index on its second coordinate derivative (KCl solution): holographic experiment data, obtained in (a) 5 min 20 s and (b) 100 min 44 s after the beginning of the diffusion process, and the data of the experiment based on the BOS method, obtained in (c) 44 min and (d) 84 min after the beginning of this process.

characteristics, revealing its properties in different representations.

It is important that in holographic double-exposure interferometry the high-precision experimental setup is necessary, including substantial protection against vibrations, coherent light source, precision unit of the special reference wave formation and high-quality optical elements. On the contrary, the correlation method allows the use of a simple experimental configuration. What is needed is just a flat random pattern fabricated by means of a common printer and a digital image recorder (a primitive digital camera is sufficient). As a source of radiation any lamp providing enough illumination for normal exposure may be used. In principle, even the rigid fixation of the camera is not required, although mounting of the camera on the same base with the background pattern and the stage with the cell eliminates additional difficulties in the processing of the resulting images.

13. Thus, using the correlation method of image comparison we have studied nonstationary refraction processes by the example of the interdiffusion of two liquids. Practical coincidence of the results of the correlation analysis with those of holographic double-exposure interferometry is demonstrated. It is shown that the correlation method may be used to determine the diffusion coefficient with good enough accuracy. The experimental implementation of the BOS method does not require precision setups, special devices and coherent radiation sources.

References

1. Borkova V.N., Kraiskii A.V., Mironova T.V., Sultanov T.T. *Kratk. Soobshch. Fiz. FIAN*, (7), 38 (2006).
2. Kraiskii A.V., Mironova T.V. *Kratk. Soobshch. Fiz. FIAN*, (8), 14 (2008) [*Bulletin of the Lebedev Physics Institute*, **35**, 231 (2008)].
3. Kraiskii A.V., Mironova T.V. *Izmer. Tekhn.*, (5), 26 (2011) [*Measurement Techniques*, **54** (5), 511 (2011)].
4. Kraiskii A.V. Kudryavtsev E.M., Mironova T.V., Sultanov T.T. *Kratk. Soobshch. Fiz. FIAN*, (9), 18 (2012) [*Bulletin of the Lebedev Physics Institute*, **39** (9), 257 (2012)].
5. Kraiskii A.V. Preprint FIAN No. 222 (Moscow, 1988); http://preprints.lebedev.ru/?p=784.
6. BorkovaV.N., Zubov V.A., Kraiskii A.V. Preprint FIAN No. 5 (Moscow, 1989); http://preprints.lebedev.ru/?p=786.
7. Borkova V.N., Zubov V.A., Krajskij A.V. *J. Sov. Laser Research*, **11** (4), 305 (1990).
8. BorkovaV.N., Zubov V.A., Kraiskii A.V. *Trudy FIAN*, **212**, 58 (1991); http://proceedings.lebedev.ru/212.
9. Fink L.M. *Signaly, pomekhi, oshibki. Zametki o nekotorykh neozhidannostyakh, paradoksakh i zabluzhdeniyakh teorii svyazi* (Signals, Noises, Errors. Notes on Some Unexpectedness, Paradoxes and Mistakes of the Theory of Communication) (Moscow: Radio i Svyaz', 1984).
10. BorkovaV.N., Zubov V.A., Kraiskii A.V. *Opt. Spektrosk.*, **63** (2), 384 (1987) [*Opt. Spectrosc.*, **63** (2), 224 (1987)].
11. Mironova T.V. *Avtometriya*, (5), 84 (2014)[*Optoelectronics, Instrumentation and Data Processing*, **50** (5), 498 (2014)].
12. Kikoin I.K. (Ed.) *Tablitsy fizicheskikh velichin. Spravochnik* (Handbook of Tables of Physical Quantities) (Moscow: Atomizdat, 1976).
13. Yakovlev K.P. *Fizicheskii praktikum* (Book of Physical Problems) (Moscow-Leningrad: Gostekhizdat, 1949) p. 38.