Diffusion study by a holographic method

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We describe the results of a study of diffusion by a holographic interferometry method with separated exposure times, and present the theory of the new method.

At the present time, holographic interferometry is finding more and more applications as a sensitive method that makes it possible, at the accuracy inherent in optical interferometry, to measure small temporal variations of the investigated object (study of deformations, vibrations, gas streams, etc.).

We have applied this method to the investigation of the diffusion process in liquids. Figure 1 shows a diagram of the employed setup. Light from a helium-neon laser passes through an optical system that broadens the light beam; the system consists of a microobjective O_1, and objective O_2, and a filtering diaphragm D (~0.03 mm diameter). The light beam reflected from the beam-splitting plate P_1 produces an object wave passing through the glass cell C with the investigated liquid. The light-splitting plate P_2 and the mirror M_1 serve to equalize the optical paths traversed by the reference and object waves from the light source to the photographic plate Ph. The light beam passing through the splitting plate P_1 and reflected from the mirror M_2 produces the reference wave. A cell with plane-parallel walls is filled with two liquids having refractive indices n_1 (in the lower half of the vessel) and n_2 (in the upper half).

The gist of the method proposed for measuring the diffusion coefficient consists in the following: At the initial instant of time, after filling the cell, the interface between the liquids is abrupt (the dashed straight lines in Fig. 2a). In the course of time this boundary becomes smeared out as a result of the diffusion, and the gradient of the refractive index decreases. Assume that after t seconds following the start of the diffusion the distribution of the refractive index over the height of the cell is described by curve I of Fig. 2a (it is assumed that the interface is located at half the height h of the cell). After a time interval Δt following this instant (i.e., $t+\Delta t$ after the start of the diffusion), the refractive-index gradient still decreases and its distributions over the height of the cell can be represented by curve II of Fig. 2a.

A hologram of the light wave that passed through the cell at the instant of time is photographed on a plate, and the hologram of the light wave at the instant of time $t+\Delta t$ is then photographed on the same plate.

The reconstructed real image of the cell will reveal alternating horizontal dark and light interference fringes. The dark fringes will be seen in those cell-image places (at those values of x) where the change of the refractive index in the interval of time between two exposures satisfies the condition $d\Delta n=(2k+1)\lambda/2$ (d is the cell thickness, λ is the wavelength of the light, and k is an integer).

The distribution of the interference fringes in the image of the cell makes it possible, in principle, to determine the diffusion coefficient. However, to increase the accuracy of the method, the interference patterns are photographed in the following sequence. The first exposure is taken at the instant of time t (after pouring the liquids). During the time of the exposure (~1 sec), the distribution of the refractive index in the liquid remains practically unchanged. Half of the reference beam (right or left) is then covered, and a second hologram is taken at the instant Δt after the first exposure. That part of the reference beam which was covered during the time of the second exposure is then uncovered, and the second half is covered. A third hologram is taken at a time interval δt after the second exposure. One-half of the interference pattern obtained in this manner will show the holograms taken

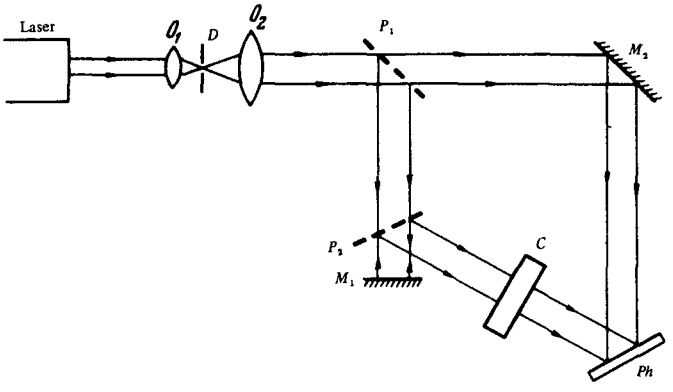

FIG. 1.

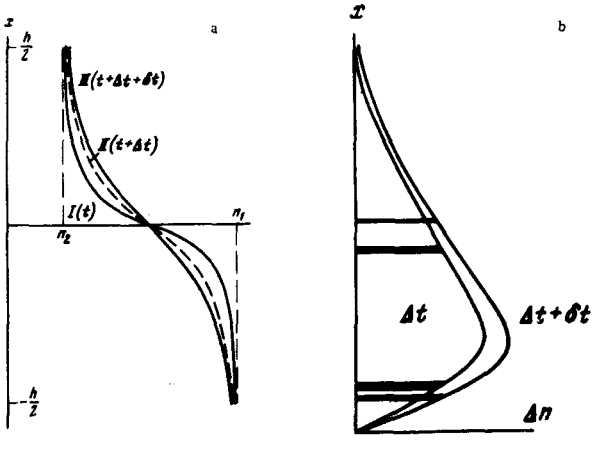

FIG. 2.

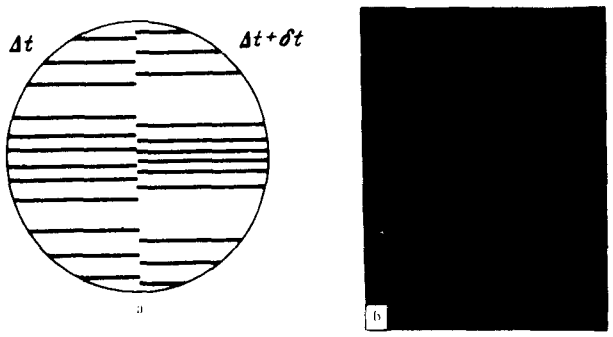

FIG. 3.

at the instants t and $t+\Delta t$, and the other half those at the instants t and $t+(\Delta t+\delta t)$.

It follows from Fig. 2a that the dependence of the difference Δn of the refractive indices on the coordinate x at a given value of the time interval between the exposures can be represented in the form of Fig. 2b (two curves are shown for two time intervals between successive exposures Δt and $\Delta t+\delta t$). The difference of the refractive indices first increases rapidly with increasing x, the order of the interference of the bands increases, and the interference fringes in this region lie close to one another. Near the maximum of Δn, a change of x results in a small change of the refractive-index difference, and the distance between the interference fringes increases. Further increase of x leads to a decrease of Δn, and accordingly to a lowering of the order of the interference.

In the reconstructed real image of the cell, the interference pattern will take the form shown schematically in Fig. 3a. The fringes located in the region where Δn increases with increasing x shift towards the center of the picture with increasing time interval between exposures. In the region where Δn decreases, the shift of the fringes is positive. This is obvious from examination of Fig. 2b, where the thick lines show the positions of two fringes for the time interval Δt, and the thin ones for $(\Delta t+\delta t)$.

The diffusion coefficient can be approximately calculated from the shift of the interference fringes in the following manner. It is assumed in the calculation that Δt and δt are much less than t and that when the time interval between exposure is increased by δt the shift δx of the fringe satisfies the relation $\delta x \ll x$. The refractive-index difference corresponding to some fringes obtained as a result of two exposures with time interval Δt between them is equal to

$$\Delta n = \frac{\partial n}{\partial t}\,\Delta t. \tag{1}$$

If a fringe of fixed order is shifted, the following condition should obviously be satisfied:

$$d(\Delta n) = \frac{\partial \Delta n}{\partial t}\,dt + \frac{\partial \Delta n}{\partial x}\,dx = 0. \tag{2}$$

Using (1), we can rewrite (2) in the form

$$\frac{\partial n}{\partial t}\,\delta t + \frac{\partial^2 n}{\partial x \partial t}\,\Delta t\,\delta x = 0. \tag{3}$$

The derivatives contained in this equation can be calculated by using the diffusion equation, which in the case of sufficiently dilute solutions can be expressed in the form

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial x^2}, \tag{4}$$

where D is the diffusion coefficient.

The solution of this equation, which determines the dependence of the refractive index on the coordinate and on the time elapsed from the instant of the start of diffusion, is

$$n(x,\ t) = \frac{n_2 - n_1}{\sqrt{\pi}} \int_{-\infty}^{\xi} e^{-\xi^2} d\xi + n_1 \tag{5}$$

where $\xi = x^2/2\sqrt{Dt}$ (see[1]).

Substituting this solution in (3) we obtain

$$D = \frac{x^2 \Delta t\,\delta x}{2t(x\delta t + \Delta t\,\delta x)}. \tag{6}$$

To calculate the diffusion coefficient one uses only fringes located far from the initial interface between the liquids, and for which the shift from the center of the picture increases with increasing time interval between successive exposures. The reason is primarily that the dependence of the refractive indices on the time is determined, for regions far from the interface, mainly by the diffusion process itself, and not by the inevitable mixing of the liquids near their interface during the time of pouring.

Owing to the refraction in the liquid, the arrangement of the interference fringes relative to the initial interface of the liquid is not quite symmetrical. To determine the coordinate of some fringe one therefore measures the distance between fringes of like order, located on opposite sides of the interface. These fringes will be located in those places of the cell image where the refractions are equal.

The described method was used to investigate the diffusion of sodium chloride in water.

Figure 3b shows a photograph of the reconstructed image of the cell. The value obtained for the diffusion coefficient is $D = (1.44 \pm 0.06) \times 10^{-5}$ cm^2/sec, and agrees well with the tabulated values.[2]

[1] A. N. Tikhonov and A. P. Samarskiĭ, Uravneniya matematicheskoi fiziki (Equations of Mathematical Physics), Nauka, Moscow, 1966.
[2] Handbook of Chemistry and Physics, 33rd Edition, Chem. Rubber Publ. Co., Cleveland, Ohio, 1951.