Investigation of the kinetics of fluctuation phenomena by a holographic method

O. A. Shustin, T. G. Chernevich, and S. A. Ivanov

Moscow State University
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Results are described of the measurement of the width of the spectral line of light scattered by concentration fluctuations in a bromoform propyl alcohol mixture. The diffusion coefficients of the investigated liquids are calculated on the basis of the obtained data.

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Holographic methods are finding ever increasing diverse applications in the invetigation of molecular processes. These applications include the study of diffusion,[1] of the Brownian motion of particles,[2] and others.

This paper is devoted to the measurement of the spectral line width of light scattered by concentration fluctuations in a bromoform + propyl alcohol mixture. The measurements were performed by a holographic method proposed by us, which made it possible to determine small spectral-line broadenings due to some molecular processes that lead to modulation of the light wave incident on the investigated object. The measurements were performed in the following manner: Two light waves are simultaneously incident on the same place on a photographic plate. The first is a randomly modulated light wave scattered by the investigated object (object wave) and the second is a plane light wave from the same laser source (reference wave), but making a certain angle with the object wave. The interference pattern obtained in this manner is not stable, and the interference fringes shift randomly in the course of time. If the exposure time of such a hologram is much shorter than the characteristic modulation time, then the interference pattern obtained on the photographic plate will have a certain good visibility. Consequently, the object wave reproduced with the aid of this hologram will be characterized by a sufficiently high diffraction efficiency. With increasing exposure time, the visibility of the interference pattern recorded on the photographic plate will decrease, and accordingly the diffraction efficiency of the obtained hologram will decrease. From the experimentally determined dependence of the diffrac-

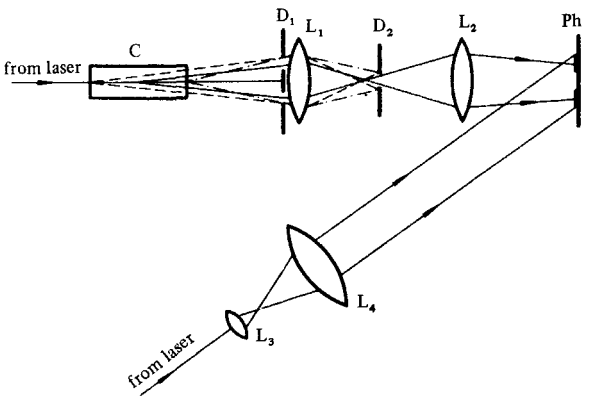

FIG. 1.

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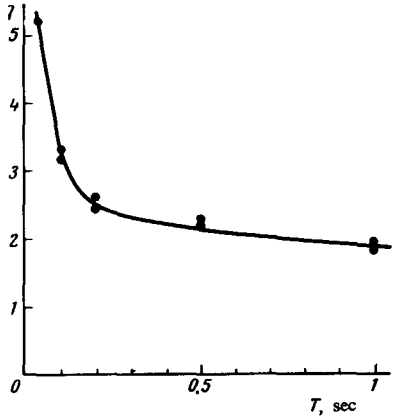

TIG. 2.

ion efficiency on the hologram exposure time it is possible, as will be shown below, to determine he width of the spectral line of the scattered light wave. The reciprocal of the spectral line width hould then lie in the region of the employed exposure times.

Figure 1 shows the experimental setup, which is assembled on a UIG-2M holographic stand. A light beam from an argon laser, model 165, is split into two. In the single-mode regime ($\lambda = 5145$ Å) the laser power was ~ 1.5 W. One beam enters the cell (C) with the scattering liquid. Behind he cell is located a system for separating the scattered light. This system consists of an annular liaphragm (D_1), a lens L_1, and a circular diaphragm (D_2). The distance from the cell to the nnular diaphragm and the diameter of the diaphragm are so chosen that light scattered at an ngle $\theta = 1.88°$ is separated. The light scattered from the central part of the cell is focused by lens $_1$ onto the center of the diaphragm D_2 (~ 1 mm diameter), and the parameters of the system are o chosen that the light scattered by the defects in the entrance and exit windows of the cell (and onsequently, by all other parts of the setup situated before and after the cell) does not pass hrough diaphragm D_2. The dashed lines in Fig. 1 show the light rays scattered from the windows f the cell. The total cone of the scattered-light rays is then incident on a photographic plate (Ph). The reference wave passing through a diverging system of lenses L_3 and L_4 is incident on the same lace on the plate. The experiment is performed in the following manner:

A series of holograms with some definite exposure time is photographed on the plate. The ntensities of the reference and object waves are varied within a small range in proportion to one nother with the aid of calibrated neutral light filters. Series of holograms with other exposure imes are then photographed on the same plate, the neutral filters being then chosen in such a way hat the densities of all the series of the holograms are approximately the same. Each hologram is a ircle of 4–5 mm diameter. After developoment, the photographic plate is placed in its previous osition and when it is illuminated by the reference wave the cone of rays of the subject wave is econstructed. The intensity of the reconstructed image is then measured for each exposure and the liffraction efficiency of the hologram is then plotted as a function of its density. From these data it s possible to obtain the dependence of the diffraction efficiency on the exposure time for a definite ıolograph density.

The points in Fig. 2 show the experimental values of the diffraction efficiency for light cattered by the two-mole solution of bromoform in propyl alcohol. The exposure time ranged rom 1/25 to 1 sec. The values of the diffraction efficiency are given in relative units.

The theoretical calculation of the dependence of the diffraction efficiency on the exposure ime was carried out in the following manner.

The reference and object waves incident on a certain section of the hologram can be represented as two plane waves in the form:

$$E_1(t) = E_{10}\left(t - \frac{\mathbf{n}_1 \mathbf{r}}{c}\right); \quad E_2(t) = f(\theta, t) E_{10}\left(t - \frac{\mathbf{n}_2 \mathbf{r}}{c} + \frac{\Delta}{c}\right).$$

Here $\mathbf{n}_1$ and $\mathbf{n}_2$ are unit vectors in the propagation direction of these waves, Δ is the optical path difference between the object and reference waves, and $f(\theta,t)$ is a modulating function whose characteristics are determined by the static and dynamic inhomogeneities of the object, as well as by the scattering angle θ. It can be assumed in addition that $f(\theta,t) \ll 1$, since the intensity of the reference wave greatly exceeds the intensity of the object wave.

The density of the section of the hologram is determined by the exposure S, which is equal to the product of the intensity of the light incident on the section of the hologram by the exposure time T:

$$S = T\{I_1 + I_2 + 2K(\tau)\,\phi(\theta,T)\},$$

where I_1 and I_2 are the intensities of the reference and object waves ($I_1 \gg I_2$),

$$I_2 = (I_1/T)\int_t^{t+T} f^2(\theta,t)\,dt; \quad \tau = \frac{(\mathbf{n}_2 - \mathbf{n}_1)\mathbf{r} + \Delta}{c};$$

$$\phi(\theta, T) = (1/T)\int_t^{t+T} f(\theta,t)\,dt;$$

and $K(\tau)$ is the autocorrelation function of the primary laser beam. At τ much less than the coherence time of the laser radiation we can assume $K(\tau) = I_1 \cos(\omega_0 \tau)$, where ω_0 is the average frequency of the laser radiation. The amplitude transmission coefficient of the hologram obtained by the method indicated above is in our case ($I_1 \gg I_2$):

$$a = A/S^{\gamma/2} \approx \frac{A}{(I_1 T)^{\gamma/2}}\left(1 - \gamma\frac{K(\tau)}{I_1}\,\phi(\theta,T)\right).$$

The constants A and γ are determined by the properties of the employed photographic material. It is known that the diffraction efficiency of a hologram with this transmission coefficient is

$$\eta(\theta,T) = \gamma^2 \langle \phi^2(\theta,T)\rangle,$$

where $\langle\phi^2(\theta,T)\rangle$ means the value of $\phi^2(\theta,T)$ averaged over the entire employed area of the hologram.

The quantity $\langle\phi^2(\theta,T)\rangle$ can be expressed in terms of the spectral intensity $G(\theta,\omega)$ of the initial process $f(\theta,t)$[3]

$$\langle \phi^2(\theta,T)\rangle = 2\int_0^\infty \left(\frac{\sin\frac{\omega T}{2}}{\frac{\omega T}{2}}\right)^2 G(\theta,\omega)\,d\omega.$$

Assuming a Lorentz line contour of the molecularly scattered light $G_L = B/[\omega^2 + \delta^2\theta]$ and taking into account the scattering by the scattering inhomogeneities of the object, we obtain ultimately:

$$\eta(\theta,T) = \gamma^2 \langle \phi^2(\theta,0)\rangle \left[\sigma + (1-\sigma)\frac{e^{-x} + x - 1}{x^2}\right], \text{ where } x = \delta(\theta)\,T.$$

$\sigma=\bar{I}_2/(\bar{I}_2+\tilde{I}_2)$, $\bar{I}_2+\tilde{I}_2=I_2$, where $\bar{I}_2$ and $\tilde{I}_2$ are the intensities of the light scattered by the static and dynamic inhomogeneities of the investigated object. Figure 2 shows a theoretical plot of the diffraction efficiency for different exposure times at a scattered-light half-width $\delta=150$ rad/sec. This result agrees, within the limits of experimental error, with Aref'ev's data and with our measurement data obtained by the square-law detection method. In the determination of the theoretical point it was assumed that the background due to the static inhomogeneities is 1.7. The error in our experiments was 100%, this being due primarily to the low sensitivity of the theoretical dependence of η on T and on the line width at the exposure times used in the present study. The accuracy can be improved by using shorter exposures.

The spectral line width of the scattered line width, determined in our experiment, has made it possible to calculate the coefficient of mutual diffusion of the investigated liquids, by using the relation[5]

$$\delta = 4n^2 Dk^2 \sin^2(\theta/2),$$

where n is the refractive index of the mixture, D is the diffusion coefficient, and $\mathbf{k}$ is the wave vector of the incident light wave. Calculation yielded $D=0.38\times10^{-5}$ cm^2 sec^{-1}.

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