Propagation of a coherent light beam through diffusing liquids and determination of the diffusion coefficient

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(Submitted July 7, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **24**, No. 3, 174–179 (5 August 1976)

We report the results of experimental and theoretical investigations of the singularities of the passage of a parallel coherent beam of light through diffusing liquids. New methods of determining the coefficient of mutual diffusion of liquids are proposed.

PACS numbers: 78.20.Dj, 66.10.+y

The refraction of incoherent light beams near the initial separation boundary of two diffusing liquids has been the subject of investigations for a long time.[1] However, experimental investigations of this phenomenon in coherent light have made it possible to observe a number of interesting features that make it possible, by comparison with the detailed theory of refraction, to determine the diffusion coefficient of the liquids by several new methods. The experimental setup is illustrated in Fig. 1. An expanded parallel laser light beam incident perpendicular to the lateral face of a cell passes through the latter. In the lower and upper parts of the cell are liquids with different densities and different refractive indices n_1 and n_2. The light-beam structure passing through the cell

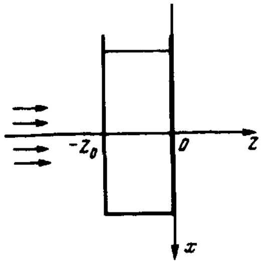

FIG. 1.

is observed on a screen or is photographed on plates placed perpendicular to the z axis (Fig. 1).

If the screen in located near the cell, then the radiation intensity distribution is uniform. At a sufficiently large distance, $z = 1.5$ m, from the cell, an interference pattern is observed, as shown in Fig. 2(a) (magnification 5×), consisting of a system of nonequidistant interference fringes parallel to the interface. The interference pattern is bounded from above and from below by brightly illuminated fringes. Each interference fringe has in turn a fine structure [Fig. 2(b), magnification 15×] consisting of a system of narrow interference fringes parallel to the interface. With increasing time after the start of the diffusion, the bright outer interference fringes approach each other and coalesce [Fig. 2(a), the position of the photographic plate on the z axis is the same for all the frames, and the time following the start of the diffusion ranges from 85 to 147 min]. The appearance of the interference pattern is due to the fact that coherent light beams passing through sections of the cell and experiencing consequently different deflections and different phase shifts arrive at a

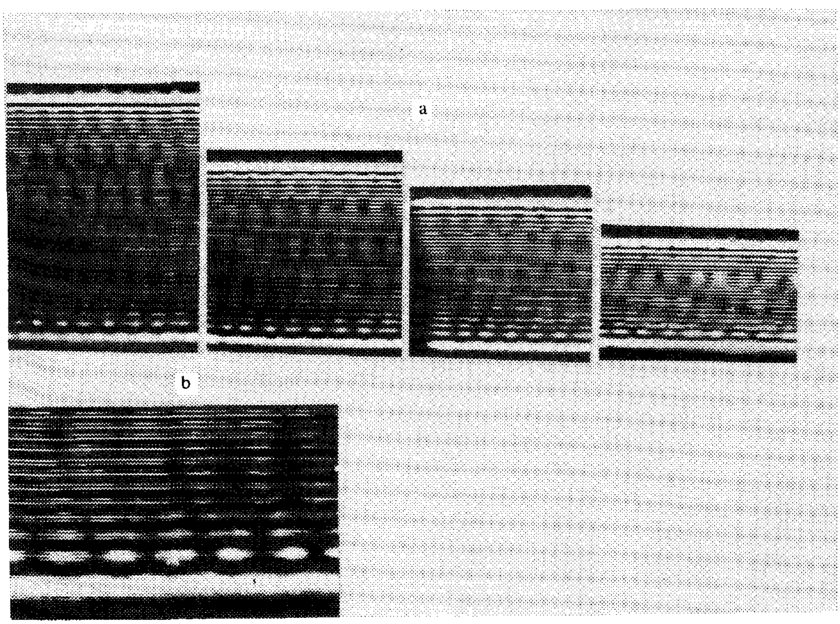

FIG. 2.

given spot on the screen. In the region where the liquids are mixed by diffusion, the refractive index is, according to[1],

$$n(x) = n_1 - \frac{n_1 - n_2}{2}\left\{1 - \phi\left(\frac{x}{2\sqrt{Dt}}\right)\right\}. \tag{1}$$

Here D is the mutual diffusion coefficient, t is the time elapsed from the start of the diffusion,

$$\phi(r_0) = (2/\sqrt{\pi})\int_0^{r_0} e^{-p^2}\,dp, \text{ and } r_0 = x/2\sqrt{Dt}.$$

A bundle of rays directed along the z axis and parallel to the separation boundary is incident on the cell (Fig. 1). In the geometric-optics approximation, the ray propagation in the cell is described by the equations

$$K_z\frac{\partial K_z}{\partial z} + K_x\frac{\partial K_z}{\partial x} = 0, \tag{2}$$

$$K_z\frac{\partial K_x}{\partial z} + K_x\frac{\partial K_x}{\partial x} = \frac{\omega^2}{c^2}n\frac{dn}{dx}. \tag{3}$$

Here K_z and K_x are the projections of the wave vector on the z and x axis. If $|n_1 - n_2| \ll n_1, n_2$, we always have $K_x \ll K_z$. Then, taking (1) into account and assuming the displacement of the ray along x in the cell to be small, we obtain from (3) on the second boundary of the cell $z = 0$:

$$K_x = \frac{\omega(n_1 - n_2)z_0}{2c\sqrt{\pi Dt}}e^{-x^2/4Dt}, \qquad K_z = K_0 = \frac{\omega}{c}n_0. \tag{4}$$

Here n_0 is the refractive index of air, z_0 is the thickness of the cell, ω is the frequency of the light wave, and c is the speed of light in vacuum. Thus, on leaving the cell, owing to refraction in the inhomogeneous liquid mixture, the rays are deflected in the vertical x direction.

The rays leaving the cell propagate in a homogeneous medium. Equation (3) for K_x then takes the form

$$K_0\frac{\partial K_x}{\partial z} + K_x\frac{\partial K_x}{\partial x} = 0, \qquad K_z \approx K_0. \tag{5}$$

The boundary condition is set at $z = 0$ and takes the form (4). The solution of (5) with boundary condition (4) is

$$K_x = \frac{\omega(n_1 - n_2)z_0}{2c\sqrt{\pi Dt}}\exp\left[-\frac{\left(x - \frac{K_x}{K_0}z\right)^2}{4Dt}\right]. \tag{6}$$

This expression determines implicitly K_x at any point x at the instant of time t.

Let $z = z_{scr}$, z_{scr} is the distance from the cell to the screen. Depending on the value of z, the plot of K_x against x, determined by formula (6), takes the form

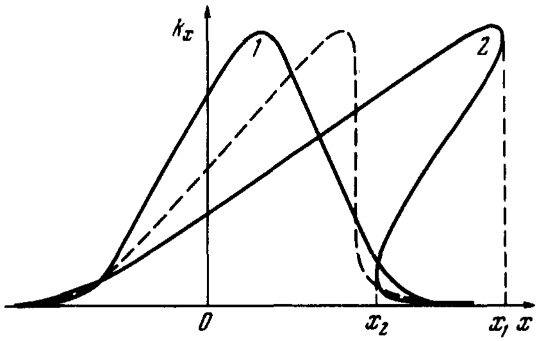

FIG. 3.

of curve 1 or 2 (Fig. 3). Curve 1 has no vertical tangents, while curve 2 has these tangents at points x_1 and x_2 corresponding to the intersection of the screen plane $z = z_{scr}$ with the caustic surface.

The condition that determines in this case the existence of caustics is

$$\partial K_x / \partial x \to \infty . \tag{7}$$

From (6) with allowance for (7) we can obtain the equation of the caustics

$$a = \tau e^{-\tau^2}, \tag{8}$$

where

$$a = \frac{2\sqrt{\pi} D t}{(n_1 - n_2) z_0 z_{scr}}, \qquad \tau = \frac{x_K - \frac{K_{x_0}}{K_0} z_{scr}}{2\sqrt{Dt}} \tag{9}$$

x_c is the coordinate of the caustic, and K_{x_0} is the value of K_x on the caustic. At $a > a_m = (1/\sqrt{2}) e^{-1/2} = 0.429$, Eq. (8) has no solutions. In this case there are no caustics. At $a < a_m$, the equation has two solutions, τ_1 and τ_2. In this case there are two caustics at two values of τ (9). Since $a \sim 1/z$, it follows that at large z_{scr} there are always two caustics, and at small z_{scr} there are no caustics. At $a = a_m$ and $\tau = \tau_m = 2^{-1/2}$ the caustics coalesce; this case is illustrated in Fig. 3 by the dashed curve.

The distribution of the field intensity near the caustics, taking into account the diffraction, is described by the expression[2]:

$$I \sim F^2(\xi); \qquad \xi = (x - x_c)/\alpha^{1/3}, \tag{10}$$

where $F(\xi)$ is an Airy function. The parameter α is connected with the caustic parameter τ by the relation

$$\alpha = \frac{z_{scr}^2}{2\sqrt{Dt} K_0^2} \left(\frac{2\tau^2 - 1}{\tau} \right). \tag{11}$$

As seen from Fig. 3, besides the two rays forming the caustics, a third ray passes through the given point x_c. The interference then leads to modulation of the intensity (10) with a period $2\pi/K_3$, where K_3 is the projection of the vector of the third ray on the x axis.

Thus, the bright light fringes that outline the interference field on Fig. 2 are the intersection of the caustics with the plane of the photographic plate. The gross structure of the fringes gives the field distribution near the caustics, while the fine structure is a consequence of interference with the third ray. A detailed comparison shows that the experimental data are in sufficiently good quantitative agreement with theory. This enables us to determine the diffusion coefficient D. We indicate three ways of doing this.

1. *From the coalescence of the caustics.* Let the caustics coalesce at the instant t_0. Then $a = a_m = (1/\sqrt{2})e^{-1/2}$. It follows therefore from (8) that

$$D = \frac{e^{-1/2}}{2^{3/2}\sqrt{\pi}} \frac{z_{scr} z_0 (n_1 - n_2)}{t_0}. \tag{12}$$

Here z_0 is the cell thickness and z_{scr} is the distance from the cell to the screen.

2. *From the positions of the caustics.* It follows from (8) and (9) that

$$\frac{x_c}{\sqrt{A_0}} = \frac{e^{-\tau^2/2}}{\sqrt{\tau}}(2\tau^2 + 1), \text{ where } A_0 = \frac{(n_1 - n_2) z_0 z_{scr}}{2\sqrt{\pi}}. \tag{13}$$

By determining from experiment the positions $x_c^{(1)}$ and $x_c^{(2)}$ of the caustics, at a given t, we obtain from (13) the corresponding values of τ_1 and τ_2 and we determine from formulas (8) and (9) the diffusion coefficient

$$D = \frac{A_0 \tau_1 e^{-\tau_1^2}}{t} = \frac{A_0 \tau_2 e^{-\tau_2^2}}{t}. \tag{14}$$

In similar fashion it is possible to determine the diffusion coefficient D also from the difference of the positions of the caustics.

3. *From the interference fringes.* Near the caustics, the distance between the minima of the field is given by

$$x_n - x_{n-1} = a^{1/3}(\beta_n - \beta_{n-1}), \tag{15}$$

where x_n and x_{n-1} are the neighboring minima of the field, and β_n and β_{n-1} are the corresponding zeroes of the Airy function. After determining a from the experimental data, we obtain from (11) the diffusion coefficient

$$D = \frac{z_{scr}^4}{4k_0^4 t} \frac{(2\tau^2 - 1)^2}{\tau^2 a^2}. \tag{16}$$

The diffusion coefficients obtained from experiment by the indicated three methods for a 10% solution of NaCl in H_2O diffusing in water turned out to be respectively $D \cdot 10^5$ cm^2/sec $= 1.29 \pm 0.02$, 1.34 ± 0.04 and 1.33 ± 0.02.

[1]A. Sommerfeld, Optics, Interscience.
[2]L. D. Landau and E. M. Lifshitz, Teoriya Polya, Nauka, 1967 [Classical Theory of Fields, Pergamon, 1971].