Measurement of the diffusion coefficient in liquids using Fresnel diffraction from a phase step

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Abstract

In this paper, we use Fresnel diffraction from a phase step in transmission to measure the diffusion coefficient in a transparent liquid. A transparent glass plate immersed vertically in a cell containing two diffusing liquids makes a phase step in transmission. When it is illuminated by a parallel beam of light, the diffraction pattern is formed on a screen perpendicular to the direction of the light beam. The diffusion of liquids inside the cell leads to the variation of the diffraction pattern along the edge of the glass plate. The refractive index gradient in the liquids is then obtained by analyzing the diffraction patterns at different times after the beginning of the diffusion process. We then get the diffusion coefficient from the measured gradient. Using this method, we study the diffusion process of the sucrose-water solution and report the diffusion coefficient with a reliable accuracy.

1 Introduction

The diffusion process is ubiquitous in nature, science, and technology, ranging from pollution control, chemical and mechanical engineering, to crystal growth, biological systems, and separation of isotopes [1–3]. Among various chemical and physical methods for studying this process in transparent media, the most important ones are the optical methods, which mainly are based on measuring the refractive index or its derivative variation [4, 5]. Interferometry methods such as Michelson interferometry [6], Mach–Zehnder interferometry [7], electronic speckle pattern interferometry (ESPI) [8], digital holographic interferometry [9], and the Moiré technique [10] have been used to study the diffusion process. Besides that the interferometry techniques are noninvasive and give real-time results, their main advantage is that they are highly accurate and reliable. There are however some drawbacks for the interferometry techniques such as their sensitivity to mechanical noises, their experimental setup complications, and the difficulties in the measuring procedures.

A widely used method for deriving the diffusion coefficient is determining the distance between the peaks in concentration difference profile between two times. Also, some other methods including tracing the interference fringes and fitting the concentration profile equations to the experimental data at different times have been developed [9].

Discontinuous changes in phase or phase gradient of the light beam lead to appreciable diffraction referred to as Fresnel diffraction from phase steps [11, 12]. This effect has been studied theoretically and experimentally [13, 14] and applied to plenty of interesting metrological measurements. The measurements include refractive indices of materials with high accuracy [15–18], nonlinear refractive index [19], thickness of thin films and plates [20], nanometer displacement [21], wavelength, and spectral line profile [17, 22].

In this paper, we introduce a new method for the study of the diffusion process based on the Fresnel diffraction from a phase step in transmission. The refractive index profile of the diffusing liquids is determined using the Fresnel diffraction pattern of the edge of a glass plate immersed vertically inside a transparent cell. Then the diffusion coefficient of liquids is obtained using the refractive index profile at two different times. This method has all the advantages of the interferometry techniques, besides, practically is insensitive to vibrations and easily applicable.

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2 Theory

In Fig. 1a, a parallel beam of light strikes the edge area of a transparent glass plate of thickness \( d \) and refractive index \( n_0 \). The transmitted light forms a diffraction pattern on a screen perpendicular to the beam direction. The normalized intensity on the diffraction pattern at arbitrary point \( P \) is given by [11, 13]

\[
I_n(P) = \cos^2\left(\frac{\phi}{2}\right) + 2(C_0^2 + S_0^2)\sin^2\left(\frac{\phi}{2}\right) + (C_0 - S_0)\sin\phi,
\]

where \( \phi \) is the phase change at the plate edge, and \( C_0 \) and \( S_0 \) are the Fresnel cosine and sine integrals [23]. A typical diffraction pattern of the plate edge and the corresponding intensity profile are shown in Fig. 1b and c, respectively. Here, we consider a free diffusion process in one dimension that causes a refractive index gradient in a specified direction. The free diffusion process is characterized by a constant diffusion coefficient, \( D \), and described by Fick’s second law

\[
\frac{dC(y, t)}{dt} = D \frac{d^2C(y, t)}{dy^2},
\]

where \( C(y, t) \) is the concentration at position \( y \) and time \( t \). A solution to Eq. (2) for the mixture of two liquids with concentrations \( C_1 \) and \( C_2 \), initially separated at \( y = 0 \) is given by [24]

\[
C(y, t) = \frac{C_1 + C_2}{2} + \frac{C_1 - C_2}{2} \text{erf}\left(\frac{y}{2\sqrt{Dt}}\right),
\]

where \( \text{erf}(x) \) is the error function. For small concentration ranges, the relation between the concentration and the time–dependent refractive index is linear as follows [5]

\[
n(y, t) = \left(\frac{dn}{dc}\right) C(y, t) + n',
\]

where \( \left(\frac{dn}{dc}\right) \) stands for the mean slope of the refractive index curve versus concentration in the applied concentration range and \( n' \) is a constant. Therefore, we can consider the refractive index distribution inside the diffusion cell as

\[
n(y, t) = \frac{n_1 + n_2}{2} + \frac{n_1 - n_2}{2} \text{erf}\left(\frac{y}{2\sqrt{Dt}}\right),
\]

where \( n_1 \) and \( n_2 \) are the refractive indices corresponding to concentrations \( C_1 \) and \( C_2 \), respectively. Now, denoting the refractive index difference at two different times as

\[
\Delta n(y, t_2, t_1) = n(y, t_2) - n(y, t_1),
\]

and, using Eqs. (5) and (6) we get

\[
\Delta n(y, t_2, t_1) = \frac{n_1 - n_2}{2} \left(\text{erf}\left(\frac{y}{2\sqrt{Dt_2}}\right) - \text{erf}\left(\frac{y}{2\sqrt{Dt_1}}\right)\right).
\]

The time-dependent phase of the light beam passing through the edge area of the plate inside the cell is related to the refractive index as follows

\[
\phi(y, t) = \frac{2\pi}{\lambda} (n(y, t) - n_0) d,
\]

where \( \lambda \) is the wavelength of the incident light. The change of the refractive index during the diffusion process modifies the phase difference along the plate edge. Therefore, using Eq. (8), we can write

\[
\Delta\phi(y, t) = \frac{2\pi d}{\lambda} \Delta n(y, t).
\]

By derivating from Eq. (9),

\[
\Delta\phi(y, t) = \frac{2\pi d}{\lambda} n'(y, t).
\]
and solving the resultant equation, gives two local extremes as follows
\[ y_{1,2} = \pm \sqrt{\frac{2D \ln(t_2/t_1)}{(1/t_1) - (1/t_2)}}. \]  

Equation (11) reveals the relation between the extremes of the phase difference and the diffusion coefficient. Therefore, by measuring the distance between two consecutive extremes, \( \Delta y = y_2 - y_1 \), we get the diffusion coefficient from the following equation:
\[ D = \frac{\Delta y^2 \left( \frac{1}{t_1} - \frac{1}{t_2} \right)}{8 \ln t_2/t_1}. \]  

### 3 Experimental results

According to Fig. 2, a beam of light emitted from a diode laser with wavelength \( \lambda = 655 \) nm is expanded and collimated using a spatial filter (SF) and a lens (L). The beam is transmitted through a transparent rectangular cell of dimensions \( 25 \times 36 \times 36 \) mm\(^3\), DC, containing the diffusion solution. The heavier solution (3% sucrose-water solution) is introduced from below the lighter solution (distilled water) using a capillary tube to avoid any convection flow. A glass plate of thickness 2 mm, GP, is installed inside the cell, perpendicular to the beam direction. The diffraction pattern of the light diffracted from the edge of the plate is recorded by a camera (Nikon D5200, 6000 \( \times \) 4000 resolution, and 3.9 \( \mu \)m pixel pitch) every 5 min.

Figure 3a–d show the diffraction patterns at times, \( t = 30, 60, 100 \) and 150 min after the beginning of the diffusion process. The corresponding intensity profiles of the diffraction patterns along the edge of the plate are plotted in Fig. 4a–d. According to the plots, the intensity distribution along the edge is almost periodic and the fringe pitch increases with lapse of diffusion time, until the diffusion process stops at the end of the cell. In this case the number of diffraction fringes remains constant at the recorded pattern. So, it provides the initial assumptions to use of Eq. (3).
For points on the edge line, \( x = 0 \), Eq. (1) reduced to

\[
I_n(y, t) = \frac{1}{2}(1 + \cos \phi(y, t)),
\]

where the phase difference varies with \( y \) and \( t \). According to the theoretical considerations, for determining the diffusion coefficient, we need to evaluate the phase, \( \phi(y, t) \). For this purpose, we applied the Fourier transform method through the fast Fourier transform (FFT) algorithm [25, 26]. Figure 5 shows the unwrapped phase profiles that correspond to the intensity profiles in Fig. 4a and c, which have been obtained at times 30 and 100 min after the beginning of the diffusion process. Subtracting the phase profiles at these times gives the phase difference curve, as illustrated in Fig. 6. Measuring the distance between the consecutive extremes on the phase difference curve gives the diffusion coefficient, according to the Eq. (12). By the way, we calculated the diffusion coefficient for several time intervals which is given in Table 1.

| \( t_1 \) (min) | \( t_2 \) (min) | \( D \times 10^5 \) (cm\(^2\)s\(^{-1}\)) | \( D_{\text{mean}} \times 10^5 \) (cm\(^2\)s\(^{-1}\)) |
|-----------|-----------|----------------|----------------|
| 30        | 60        | 0.54           | 0.54 ± 0.03     |
| 40        | 70        | 0.50           |                |
| 50        | 80        | 0.52           |                |
| 30        | 70        | 0.54           |                |
| 60        | 100       | 0.57           |                |
| 40        | 90        | 0.52           |                |
| 40        | 100       | 0.54           |                |
| 40        | 110       | 0.55           |                |
| 30        | 110       | 0.57           |                |

The obtained mean value is \((0.54 ± 0.03) \times 10^{-5} \text{ cm}^2\text{s}^{-1}\), which is comparable with the standard value of \(0.52 \times 10^{-5} \text{ cm}^2\text{s}^{-1}\) for low concentrations at 25 °C [27]. It should be noted that the uncertainty of the diffusion coefficient measurement is derived by calculating the standard deviation of the measured values in Table 1.

4 Conclusion

Using the Fresnel diffraction from a phase step in transmission, a method is introduced for measuring the diffusion coefficient in transparent solutions. This method compared with the corresponding techniques in the field such as interferometry methods is considerably simple and insensitive to the environmental vibrations. It is suitable for a wide range of concentrations. Like other optical methods, it is limited to transparent liquids. In a case that one deals with the solutions with low concentration differences, we propose to use a wedge instead of the plate and analyze the diffraction fringes.

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