

Scanning beam technique to measure small diffusion coefficients

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In order to measure small diffusion coefficients of miscible fluids, the deflected beam technique was improved by introducing a method whereby deflections corresponding to different positions of the laser beam were compared. The method requires only knowledge of the vertical position difference Δz of the laser beam at different times, without any requirement for an assignment of the initial vertical coordinate of the fluid interface.

1. Introduction

In studying hydrodynamic instabilities between two miscible fluids [1,2] one often faces the problem of assigning a reliable value to very small diffusion coefficients (of the order of $10^{-6} \text{ cm}^2 \text{ s}^{-1}$). As for diffusion between two binary mixtures such as water and glycerin, water and salt, etc., no complete data are available in the literature. In this paper, we present an improved version of the light beam deflection technique [3,4], using a scanning laser beam and a position sensitive detector PSD to measure small diffusion coefficients of such binary mixtures, that yields fast results.

2. Theoretical background

Let us consider how two miscible fluids A and B superposed vertically in a cell diffuse in course of time. At time $t=0$, we deposit a thick layer of B on top of A, at the coordinates $z=0$. To avoid a Rayleigh-Taylor instability, the lighter liquid B is put over the heavier one A and we put a cover on the cell to avoid evaporation of the upper component, and consequent thermal gradients.

Figure 1a shows the evolution of the separation

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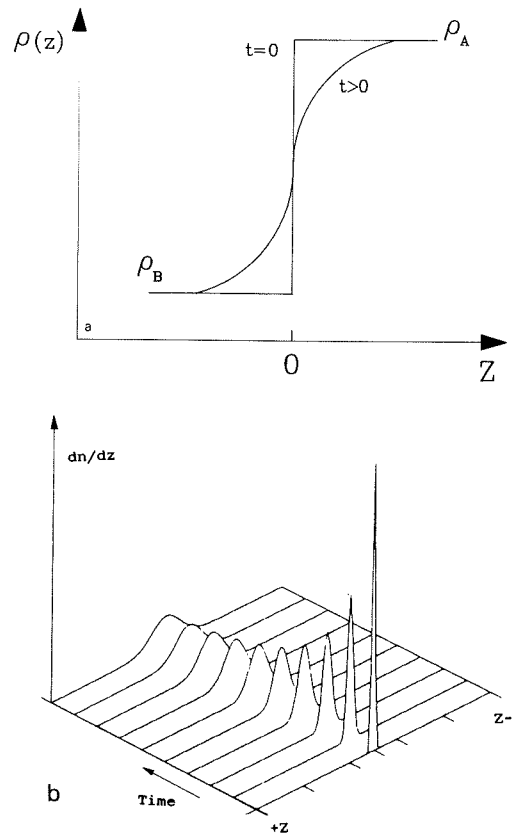


Fig. 1. (a) Time evolution of density ρ , between two miscible fluids, in the separation front ($z=0$). (b) Schematic view of the vertical gradient of refractive index.

front. For $t > 0$, the density field $\rho(z)$ evolves as sketched in the figure, reaching the values ρ_A and ρ_B (that is, the densities of the unmixed fluids) for $z \rightarrow \mp \infty$, respectively. $C(z)$ obeys a diffusion equation given by [5,6]

$$\partial C / \partial t = D \partial^2 \rho / \partial z^2, \tag{1}$$

where D is the diffusion coefficient that we have to measure. The solution of eq. (1) with the appropriate boundary and initial conditions yields

$$\rho(z, t) = \frac{1}{2}(\rho_A + \rho_B) - \frac{1}{2}(\rho_A - \rho_B) \operatorname{erf}(z / \sqrt{4Dt}), \tag{2}$$

where the error function is defined as [7]

$$\operatorname{erf}(z / \sqrt{4Dt}) = 2\sqrt{4Dt} \int_0^z \exp(-z' / \sqrt{4Dt}) dz'. \tag{3}$$

To perform an optical measurement, we rely on the fact that the refractive index $n(z)$ is proportional to the local density (apart from a Clausius-Mossotti or Lorentz-Lorenz correction [8] that we neglect). In fig. 1b, we have plotted the vertical gradient of the refractive index, which evolves as

$$\frac{dn}{dz} = \frac{\Delta n}{\sqrt{\pi 4Dt}} \exp(-z^2 / 4Dt), \tag{4}$$

where $\Delta n = n_B - n_A$ is the difference of the refractive indices of the two fluids.

Some techniques make use of a test cell which is part of an interferometer [5,9,10]. Other techniques use moiré pattern generated by two gratings placed in the opposite wall of a cell [11,12]. The fringe distribution at a given time t is subtracted from the fixed distribution at a later time $t_2 = t_1 + \Delta t$; since the phase distribution, which behaves like the refractive index, is proportional to eq. (2).

Preliminary tentatives were made using a commercial Fizeau interferometer (ZYGO). In our case the difference Δn between the refractive indices of the two liquids (a binary mixture of water and glycerin) is very strong, consequently the fringes are too dense to be resolved at the interface, so the accuracy in measuring D is not satisfactory. For this reason, we prefer to use the light beam deflection technique which is more accurate in the case of strong difference of n .

Similar considerations apply to the use of the grid technique recently introduced by Gurfein et al. [13]. Both fringe and grid methods are very good where a high sensitivity is required, however they are limited to small gradients, whereas in our case we have to deal with gradients as high as $dn/dz = 0.1 \text{ cm}^{-1}$, as can be inferred from the data reported in the following figures.

3. The deflection technique

In fig. 2 we show the principles of the deflection technique. Due to the gradient field dn/dz , a horizontal laser beam impinging the cell at $z = z_1$ is bent downwards so that the exit angle is given by [4]

$$\vartheta = l(dn/dz), \tag{6}$$

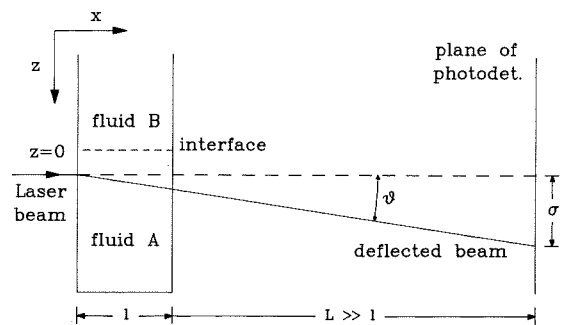


Fig. 2. Parameters in the light beam deflection technique.

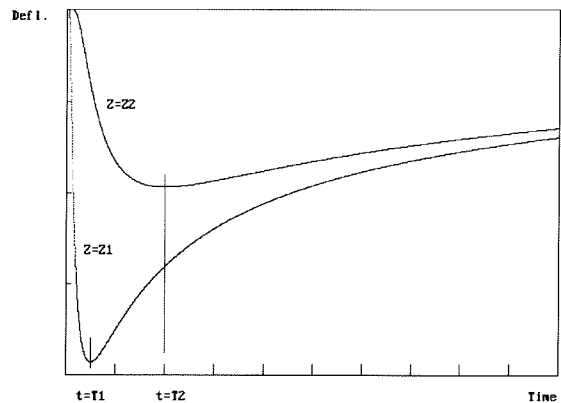


Fig. 3. Theoretical representation of the laser beam deflection versus time, for two different points of incidence Z_1 and Z_2 . The two linear scales (horizontal and vertical) are related via eq. (8).

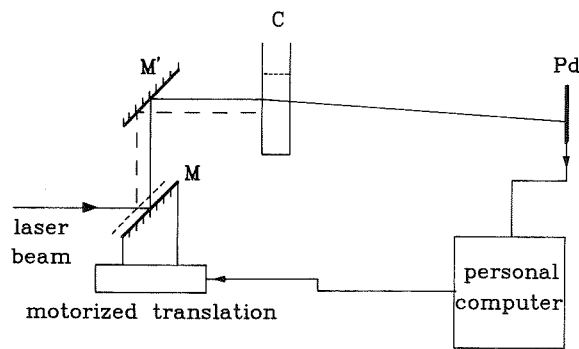


Fig. 4. Experimental set-up.

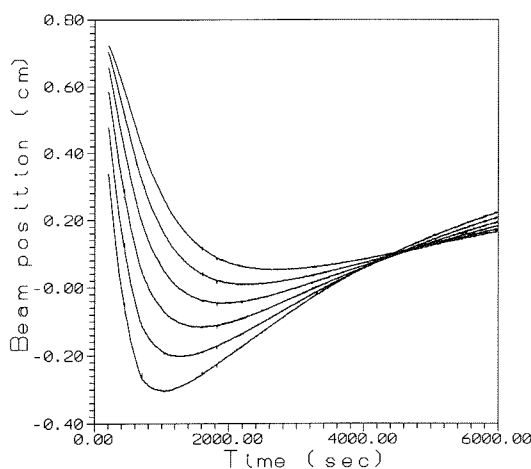


Fig. 5. Laser beam deflection measured with a position sensor detector for two mixtures A and B. (A: 40% glycerin, 60% water; B: 25% glycerin, 65% water).

where l is the cell thickness.

If we put a screen at a distance L from the cell, the vertical shift δ is given by

$$\delta = L\vartheta,$$

$$\delta(t) = Ll \frac{\Delta n}{\sqrt{\pi 4Dt}} \exp(-z^2/4Dt). \quad (7)$$

Varying the time in eq. (7), at fixed z , looks as in fig. 3. For a beam position z_1 the corresponding maximum occurs at

$$t_1 = z_1^2/2D. \quad (8)$$

Thus, measuring the time of the maximum deflection should give the value of D , provided the separation of the beam from the initial boundary of the two fluids ($z=0$) is well known.

However, in actual measurements, we fill the cell with the two fluids with very smooth operation to avoid any mixing, and then position the beam. In the process, we are unable to measure z_1 . As shown in fig. 3, we perform two measurements with poor knowledge of z_1 , and z_2 , but with a very accurate calibration of the separation $\Delta z = z_1 - z_2$, between the two measurement beam positions. By the use of eq. (8), we realise that D is given by

$$D = \frac{1}{2} \left(\frac{\Delta z}{\sqrt{t_1} - \sqrt{t_2}} \right)^2. \quad (9)$$

In practice for D values of the order of 10^{-6} , we can easily check for the first minimum, then reposition the beam by $\Delta z \approx 0.1$ cm and wait still before the second maximum appears.

4. The experimental techniques

To improve the precision of the value of D it is convenient to use a multiple beam arrangement as shown in fig. 4. The two liquids at room temp. (20°C) are stratified in a spectrophotometric glass cell C with an optical path of 1 cm. The light beam from a HeNe laser is shifted by a mirror M driven by a computer controlled translation; the light beam is moved with step Δz of 0.125 mm for a total path of 0.625 mm. For each step the positions of the deflected beam are recorded, for a scanning time of about 0.5 s, which is negligible in comparison with the time evolution of the phenomenon. Pd is a detector (lateral photoelectric effect photodiode) sensitive to the position of the centroid of the light beam [14,15]. This technique does not depend on the thickness l of the cell or the distance L from the cell to the PSD. However, high accuracy is required for measuring the time and Δz . Moreover, we can employ a low cost detector with a bad degree of linearity, because the recording of the phenomenon is relative only to the time evolution and not to the amount of the deflection.

Figure 5 shows the trend of the deflected beam position by a binary (25/40) mixture of water and glycerine. By (25/40) we mean that B has 25% of

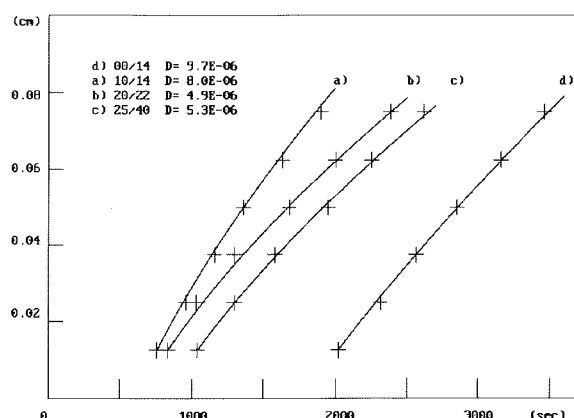


Fig. 6. Values of the time t_m corresponding to the maximum deflections for different values of Δz . Each curve is relative to different binary mixtures of water and glycerine.

heavier fluid which is glycerin and 65% of water whereas the A has 40% of glycerin and 60% of water. Each curve, relative to different values of Δz , is identified with a time t_m corresponding to the maximum deflection.

Figure 6 shows the values of the time t_m corresponding to the position of each scan, for different binary mixtures of water and glycerine. The continuous lines are obtained from eq. (9) and are relative to the best fit of D .

5. Conclusions

The standard light deflection technique requires accurate knowledge of the coordinate z of the interface separating initially the two phases. The method illustrated here relies only on knowledge of the differences Δz of the laser position at different times,

which are much more accurate than the absolute measurement of z , since they are read as positions of a motorized translator. We have carefully checked by visual inspection that no artifacts, as e.g. sedimentation, perturb our measurements. Indeed we have applied the method to completely miscible fluids.

The method presented here is particularly convenient in dealing with specimens of biological or pharmacological interest.

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