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Accuracy of the differential-interferometric measurements of curvature

Experimental study with liquid drops

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Abstract. The radius of curvature at the top of small liquid drops has been determined both visually and by means of differential interferometry in reflected light (microscope Epival Interphako, Carl Zeiss–Jena). Systematic errors in the interferometric measurements have not been found within the investigated interval of curvatures. A comparison between the data for aqueous and mercury drops showed that there is no pronounced effect of the reflectivity of the surface on the accuracy of the interferometric measurements.

1. Introduction

The differential interferometry in reflected light is a widespread and powerful method for studying the structure of solid surfaces and biological objects [1–4]. During the last decade differential interferometry (more precisely, the 'shearing method') has been applied also to fluid interfaces. Zorin [5] determined in transmitted light the profile of a biconcave liquid meniscus. Del Cerro and Jameson [6] and Mingins and Nikolov [7] measured the curvature of small oil lenses floating on an aqueous surface. The curvature at the top of spherical films of small air bubbles attached to a liquid surface has been measured [8] in reflected light in order to determine the film and line tensions [9, 10]. The application of differential interferometry to capillary phenomena was stimulated mainly by the growing interest in line tension. This quantity, which can play a significant role in the occurrence of a number of processes of practical importance [11] is usually small. Therefore its determination can be strongly affected by inaccuracies in the optical method used. Hence it is important to study possible systematic errors involved in the differential-interferometric measurement of curvature.

The differential-interferometric measurements carried out by Nikolov *et al.* [8], which provided some interesting and unexpected data on line tension, were performed by using an Epival Interphako (Carl Zeiss–Jena) microscope [12, 4]. The purpose of the present work is to check the accuracy of the optical method used in [8]. Beyer has pointed out [4] that a systematic error could appear owing to the aperture angle of the objective, and he derived theoretically the respective corrections. It is possible that some other sources of systematic errors could also exist. Hence, we undertook an experimental verification of the interferometric method using objects

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with known radii of curvature of about 100–300 μm (radii of same order were measured in [8]). It is very difficult to produce perfect solid spheres of this size, and for that reason, we preferred to use liquid drops. These surfaces are optically smooth, but deformation due to gravity must be taken into account.

2. Equations for the shape of a sessile drop

The cross-section of a sessile liquid drop on a horizontal hydrophobic surface is sketched in figure 1. The normal to the generatrix of the drop surface forms an angle ϕ with the positive direction of the z-axis. By observing the drop from the top, one can measure its equatorial radius R (at $\phi=90°$). The deviation from the spherical shape due to gravity is more pronounced for the lower drop surface ($\phi<90°$). The upper drop surface ($90°<\phi<180°$) for small drops ($R<1$ mm) is nearly spherical; more precisely it is tangent to a sphere of radius b ($b\neq R$) at $\phi=180°$. This radius b can be calculated from the measured equatorial radius R (see equation (3) below). On the other hand, the radius b can be independently measured by means of the interferometric method used in [8] and thus the accuracy of this method can be checked. To put this idea into practice we first need some equations describing the shape of such a drop.

The generatrix $z(x)$ of the drop surface satisfies the Laplace equation. Using the angle ϕ as a parameter, one can write this equation in the form [13, 14]

$$\left.\begin{aligned} d(\sin\phi)/d\bar{x}+\sin\phi/\bar{x}&=2+\beta\bar{z},\\ d\bar{z}/d\bar{x}&=-\tan\phi,\end{aligned}\right\} \tag{1}$$

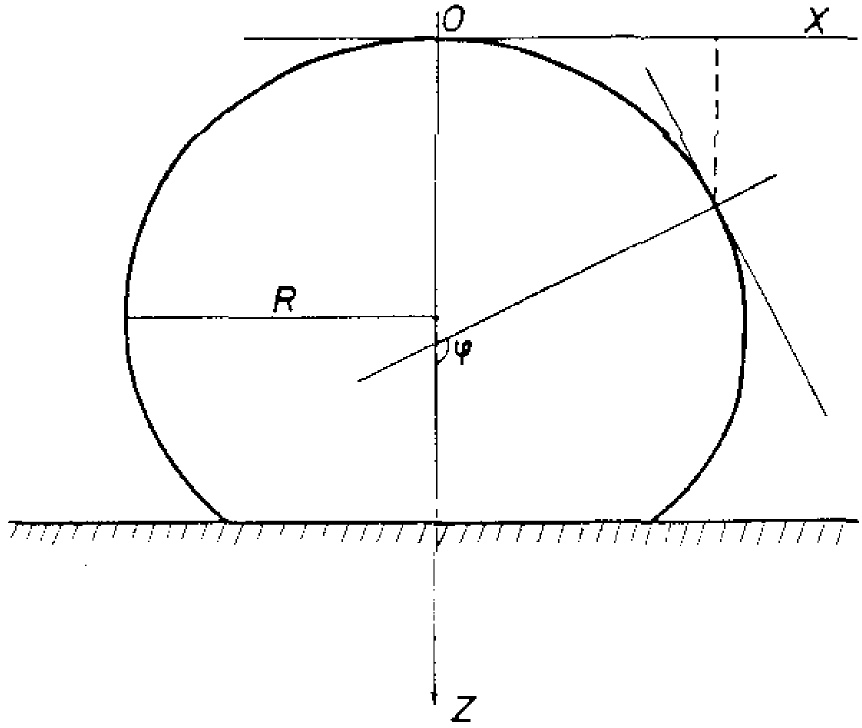


Figure 1. A sessile liquid drop on a horizontal hydrophobic surface. ϕ is the varying angle between the normal to the drop surface and the positive direction of the z-axis; R is the equatorial drop radius.

where the dimensionless variables

$$\bar{x}=x/b, \quad \bar{z}=z/b \quad \text{and} \quad \beta=\rho g b^2/\sigma \tag{2}$$

are introduced; g is the gravitational acceleration, ρ and σ are the mass density and the surface tension of the liquid, respectively. For small drops (like those considered here) β is a small parameter and the solution of equations (1) can be sought as a series expansion in powers of β. In this way a full set of asymptotic formulae for the shape of a sessile drop were derived in [15]. We will utilize some of them here for our present purpose:

(i) The radius b can be calculated from the measured equatorial radius R by means of the following equation [15]:

$$b^{-1}=R^{-1}[1-\beta/6+\beta^2(\ln 2-1/6)/6]. \tag{3}$$

Equation (3) is solved by an interactive procedure using $b^{(0)}=R$ as a zeroth approximation for b on the right-hand side of (3).

(ii) The value of the varying slope angle ϕ can be calculated for each given value of x from the equation [15]:

$$\sin(180^\circ-\phi)=\bar{x}\left\{1+\frac{\beta}{6v}\left[v-2(v-1)^2\right]+\frac{\beta^2}{6}\left(\frac{2}{v}-\ln\frac{2}{v}-1\right)\right\}, \quad 90^\circ<\phi<180^\circ, \tag{4}$$

where $v=1+\sqrt{(1-\bar{x}^2)}$.

(iii) Then with the value of ϕ so obtained the corresponding value of $\bar{z}(\bar{x})$ can be calculated:

$$\bar{z}=1+\cos\phi+\beta\{\tfrac{1}{3}\sin^2\phi+\tfrac{2}{3}\ln\sin(\phi/2)-\tfrac{1}{2}(1+\cos\phi)\}. \tag{5}$$

In other words, equations (4) and (5) give the function $\bar{z}(\bar{x})$ in an explicit (but approximate) form. In the table the accuracy of equations (4) and (5) is checked against the exact values of $\bar{z}(\bar{x})$ by computer calculations by Hartland and Hartley [14] at $\phi=175^\circ$ (near the top of the drop). In our experiments ($\beta<0{\cdot}014$, see below) $\phi>175^\circ$ so that the accuracy of the asymptotic formulae (4) and (5) is even better than that given in the table (see [15]), i.e. (4) and (5) yield practically exact values of $\bar{z}(\bar{x})$. The ratio R/b in the last column of the table is close to unity, which means that the shape of the upper drop surface is practically spherical.

Values of $\bar{z}$ calculated from equations (4) and (5) against the corresponding exact computer data z_{HH} from [14] at $\phi=175^\circ$ and different β. The ratio R/b is calculated from equation (3).

$\log_{10}\beta$	$\bar{x}$ ($\times 10^2$)	$\bar{z}$ ($\times 10^3$)	z_{HH} ($\times 10^3$)	R/b
−2·4	8·71553	3·8054	3·8053	0·9993
−2·2	8·71553	3·8055	3·8053	0·9989
−2·0	8·71549	3·8056	3·8052	0·9983
−1·8	8·71545	3·8058	3·8052	0·9974

3. Error evaluation of the interferometric method

The basic principle of the shearing method consists in splitting the original light beam, coming from the subject, into two beams, which interfere. In the focal plane of the objective one can observe two equivalent and overlapping images of the subject, displaced by a distance d, and the created interference pattern. The upper part of figure 2 is a sketch of the cross-section (in the plane $x0z$) of the two images of the reflecting surface (a sessile drop) separated at a distance d along the x-axis. The lower part of the figure is a sketch of the resulting interference pattern (in monochromatic light). The fringes, which look like parallel lines, are analogous to the so-called 'streaks' in [8]. In fact, all the fringes are loci of points for which the distance between the reflecting surfaces satisfies the following requirement:

$$|z(x+d/2)-z(x-d/2)|=n\lambda/4, \quad n=0,1,2,\ldots; \tag{6}$$

here λ is the wavelength of the light, n is the order of interference and $z(x)$ is the equation of the generatrix of the drop surface (cf. figure 1). Let us denote the right-hand side of (6) by D (this quantity is directly measurable) and the theoretically expected value of D on the left-hand side of (6) by D_t:

$$D=n\lambda/4, \quad D_t=|z(x-d/2)+z(x+d/2)|. \tag{7}$$

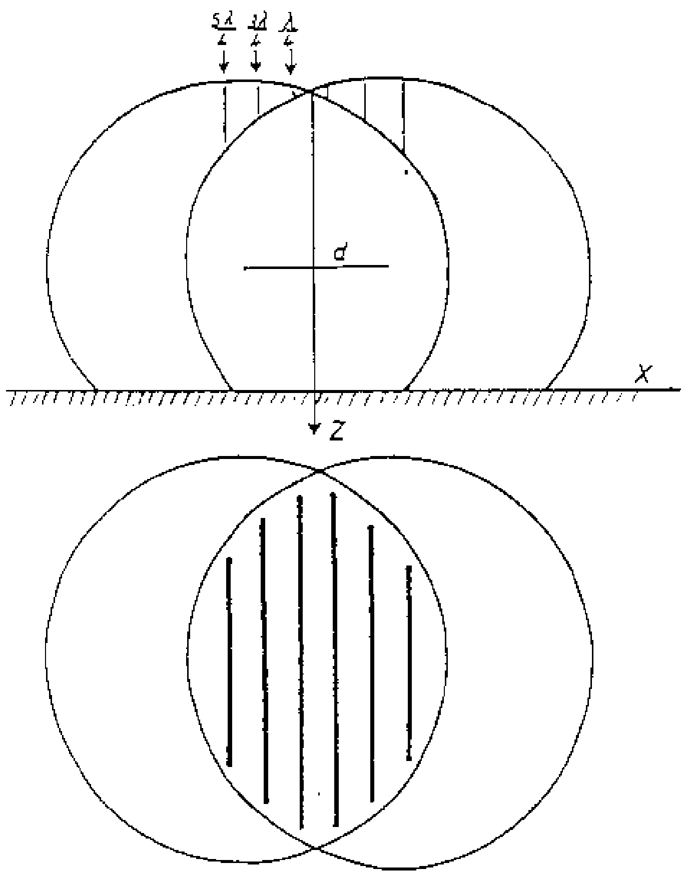


Figure 2. A sketch of the cross-section of the reflecting surfaces, shifted by a distance d (upper part) and of the resulting interference pattern (lower part).

Then, let us define the quantity

$$\delta = (D_t/D) - 1. \tag{8}$$

For each fringe one can calculate D, D_t and consequently δ. If the values of δ are predominantly positive or negative, one can conclude that a systematic error exists.

In order to estimate the inevitable statistical error in δ, we will use a suitable approximate expression for δ. It was mentioned above that in the region $175^\circ < \phi < 180^\circ$ (where the interference fringes are observed) the drop surface practically coincides with a sphere of radius b. And for this reason, in this region the generatrix of the drop surface can be approximated by

$$z(x) = \sqrt{(b^2 - x^2)}. \tag{9}$$

Taking into account that $(x \pm d/2)^2 \ll b^2$ one obtains from (7) and (9)

$$D_t \approx xd/b$$

and then from (8) one finds $\delta \approx xd/(Db) - 1$. Hence the mean-square error $\Delta\delta$ of the quantity δ can be estimated from the equation

$$\Delta\delta = \left\{ \left(\frac{d}{RD} \Delta x \right)^2 + \left(\frac{x}{RD} \Delta d \right)^2 + \left(\frac{xd}{R^2 D} \Delta R \right)^2 \right\}^{1/2}, \tag{10}$$

where we have used the fact that $b \approx R$ (cf. the table) and Δx, Δd and ΔR are the errors in the measurements of the position x of a given fringe, shearing distance d and equatorial drop radius R. The error in D is negligible because the wavelength λ is known to high precision.

4. Experimental procedure and results

The experimental set-up is shown schematically in figure 3. By ejecting distilled water from a thin syringe needle over a flat horizontal glass plate covered with Teflon tape many sessile drops of different size were formed. Owing to the hysteresis of the wetting contact angle, the equatorial cross-section of some drops were elliptical. We chose for our observations only drops of circular cross-section. The glass plate was surrounded by filter paper soaked with distilled water. In this way evaporation from the drops was reduced. The dependence of the equatorial radius of such a drop on time is shown in figure 4. The correlation coefficient of the straight line is 0·999. At some moments (denoted by arrows in the figure) photographs were taken of the interference pattern produced by the top of the drop. The microscope (Epival Interphako) and the 25 × objective were the same as in [8]. The shearing distance was $d = 12{\cdot}08$ μm with the multiple lighting slit No. 48 of the microscope. The light source was a mercury lamp HBO-50 (50 W) and the green line of wavelength $\lambda = 546{\cdot}1$ nm was used. The photographs were taken at the smallest possible opening of the field diaphragm and when the aperture diaphragm of the microscope was fixed around the fourth division. The exposure time was 4–5 s (film ORWO, 15 DIN). The times when the photographs were taken were recorded and the values of the equatorial radius at those times were found from the dependence $R(t)$ (see figure 4).

We studied in detail six photographs of the interference pattern from water drops on Teflon (one of them is shown in figure 5 (*a*)) and one photograph of a mercury drop on glass (figure 5 (*b*)). The mercury drop was used as a control to test for the possible influence of reflectivity of the drops on the accuracy of differential

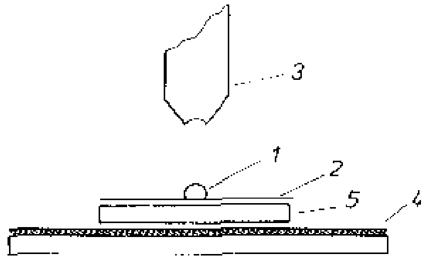


Figure 3. The experimental set-up: (1) a sessile aqueous drop; (2) Teflon tape; (3) objective of the microscope; (4) filter paper soaked with water; (5) flat glass plate.

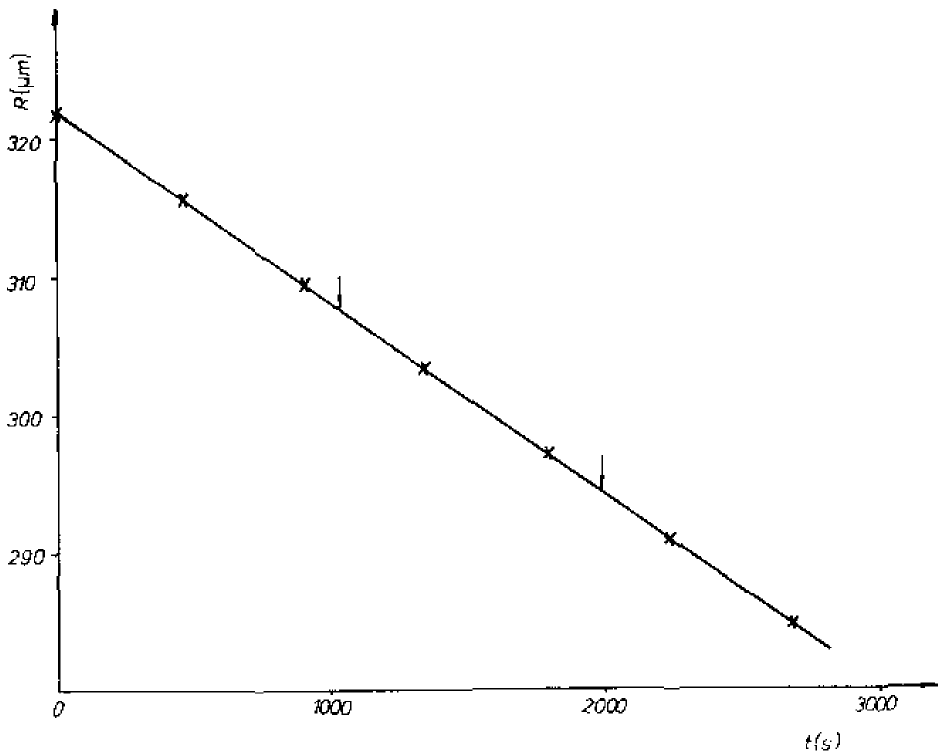
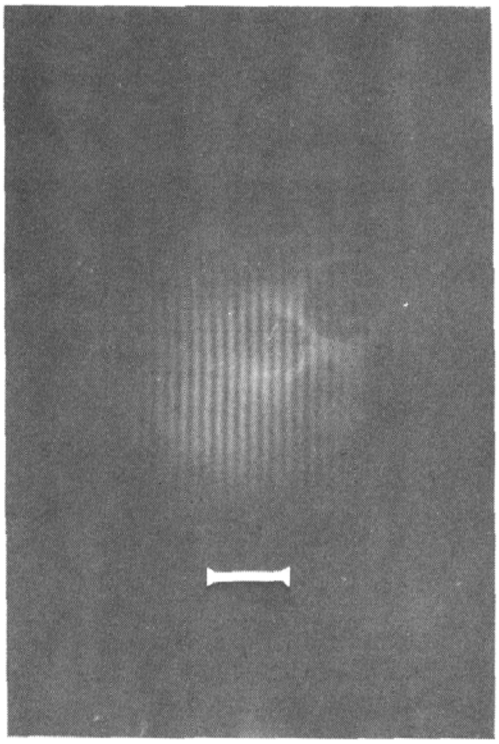


Figure 4. The decrease (caused by evaporation) of the equatorial radius R of an aqueous drop with time t. The arrows indicate the moments when photographs of the split image were taken.

interferometry. The equatorial radii of all the drops were between 190 and 310 μm. The distance x from the central fringe (see figure 2) was measured for each fringe five times.

With the measured equatorial radius R and fringe coordinate x one can calculate the value of D_l by means of equations (4), (5) and (7). Then the value of δ and its mean square error $\Delta\delta$ is found for each fringe from equations (8) and (10). The results for the seven studied drops are presented in figure 6, which shows that the quantity δ takes both positive and negative values and its mean value is close to zero. The mean-square error curve was calculated from equation (10) using experimental values



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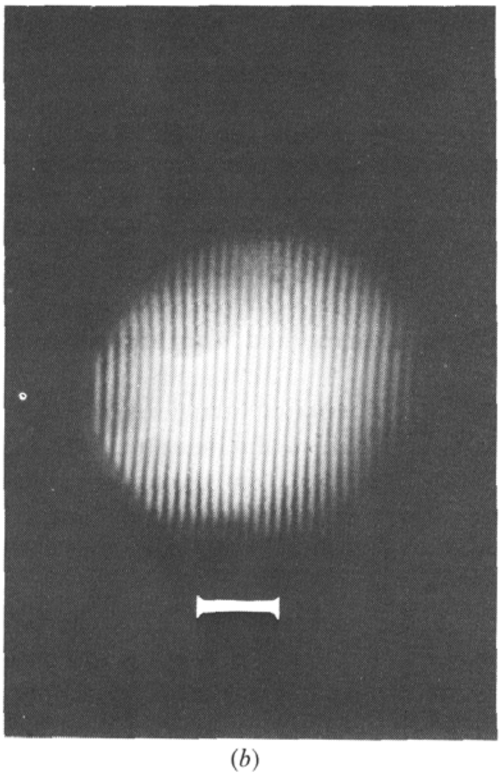

(*b*)

Figure 5. Differential interference pattern in light reflected from the top of a liquid drop: (*a*) aqueous drop ($R=308\,\mu m$), (*b*) mercury drop ($R=282\,\mu m$). Shearing distance $d=12{\cdot}08\,\mu m$; $25\times$ objective. The length of the reference marker is $50\,\mu m$.

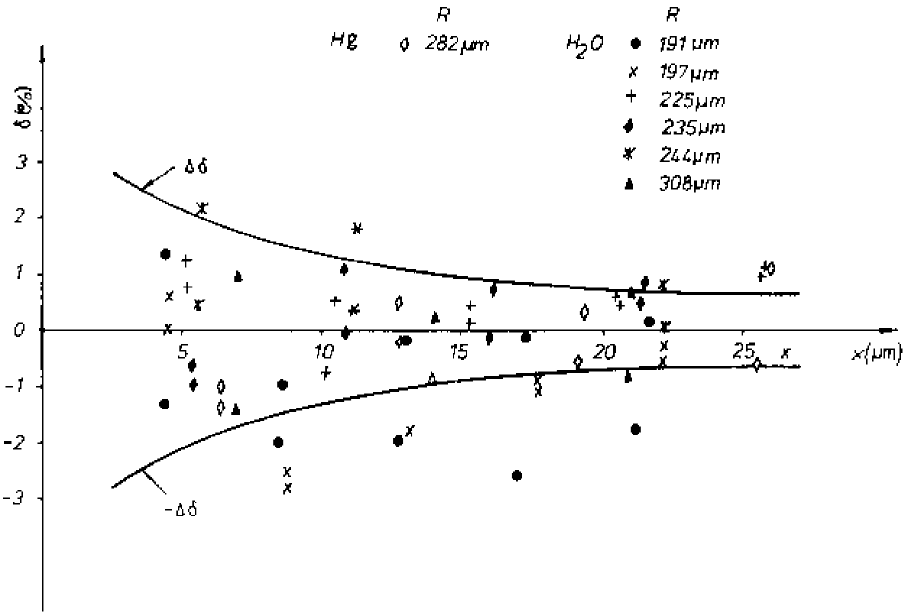

Figure 6. The quantity δ (equation (8)) versus fringe coordinate x: data from seven photographs of liquid drops. The two lines represent the standard deviation $\Delta\delta$ of δ due to random error in the measurements of x and R.

$\Delta x = 0{\cdot}11\,\mu$m, $\Delta d = 0{\cdot}02\,\mu$m and $\Delta R = 1{\cdot}5\,\mu$m. We evaluated D for each x using equation (7) for D_1. The larger drop radius $R = 308\,\mu$m was substituted in equation (10) to evaluate the lower bound $\pm\Delta\delta$.

For larger x the mean square error $\pm\Delta\delta$ is determined mainly by the random error in R. The majority of the experimental points for δ lie in the region between the two mean-square curves $\pm\Delta\delta(x)$. Hence one can conclude that the error in the optical method is due to random errors in the individual measurements. In other words, systematic error in the differential-interferometric measurement of curvature has not been found in the investigated range of curvature radii. There is no pronounced difference between the data for the mercury and water drops, and it seems that the accuracy of the optical method is not affected by the reflectivity of the surface.

We made calculations of the influence of variations in the drop surface tension σ (due to possible impurities) on the values of δ (see equations (2)–(4)). It turned out that if one varies σ from 30 to 73 mN m^{-1} for the aqueous drops and from 260 to 460 mN m^{-1} for the mercury drops, the corresponding changes in δ are of the order of 0·01 per cent, i.e. they are negligible. This is due to the very small gravity deformation of the studied small drops.

Another way of checking the accuracy of the interferometric method is to compare the results for the radius of curvature b_i at the top of the drop obtained by differential interferometry with a value of the same radius determined by an independent more direct method. Such a reference value, b_r, can be found from equation (3) by using the equatorial drop radius R, which is directly measurable. (Note that the accuracy of equation (3) is well above the accuracy of the experimental measurements.) On the other hand, equations (6), (7) and (9) can be rearranged to yield $b = (1 + d^2/D^2)\,(x^2 + D^2/4)^{1/2}$.

From the above equation and the position x of any fringe of the interference pattern one can calculate b. By taking the average of b over all fringes for a given photograph one obtains what can be called the 'interferometric value' b_i of the drop radius. In figure 7, b_i is plotted against the reference value b_r (for the same drop) for all studied drops. One sees that b_i and b_r coincide for each drop within the framework of the experimental accuracy. This fact is additional evidence that differential interferometry can be utilized for measuring curvatures (at least in the range studied by us) of fluid interfaces without any correction for systematic errors.

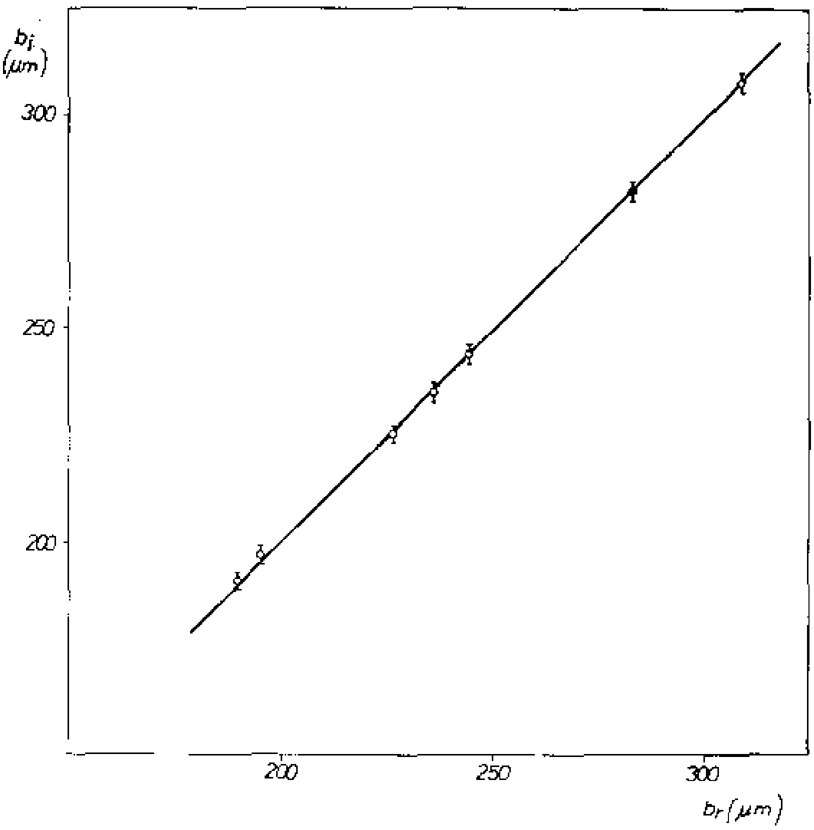

Figure 7. The interferometrically determined radius of curvature b_i versus the reference value b_r for the same drop for one mercury (●) and six aqueous (○) drops. The slope of the solid line is unity.

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