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An Optical Method for Surface Tension Measurements of Dispersed Liquid Droplets

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Abstract

An optical method for the measurement of surface tension of liquids is described. It is based on the fact that the resonance frequency of small liquid droplets depends on the surface tension. The droplet oscillation is observed with an especially designed high speed CCD camera. Preliminary results of measured surface tension values are presented.

List of Symbols

A	oscillation amplitude, << R
B,C	principal axes of spheroid
B_i,C_i	measured momentary length of axes B,C (at time t_i)
t	time
τ_n	time constant of amplitude decay
R	radius of undeformed droplet
V	volume of droplet
σ	surface tension
ν	kinematic viscosity
Ω	angular oscillation frequency
ϕ	phase angle
$\rho_{i,e}$	density of the fluid (internal and external phase)

Introduction

The knowledge of an accurate value of the surface tension for a liquid droplet is essential in a variety of two-phase processes, like the break up of liquid jets, the dispersion of one liquid in another immiscible liquid or the evaporation of dispersed liquids. It is well known that the surface tension is a very sensitive physical property and that its value depends strongly on the temperature and the purity of substances used. Hence, tabulated values of surface tension, measured under "clean" conditions, are often worthless in real processes.

The purpose of the present work is to develop a nonintrusive method to measure the surface tension of dispersed droplets in "real time" of the experimental conditions. The principle of the method is based on the observation of free oscillations of a droplet.

Oscillation Phenomena

In the absence of external forces, a liquid droplet assumes a static shape that is spherical. Any slight distortion of the shape of the droplet may involve a series of damped oscillations while returning to its former spherical shape. For a given initial state, two quantities are sufficient to completely describe the transient behavior of the particular mode of drop oscillation. These are the oscillation frequency and the amplitude decay rate, both depending on the physical properties of the liquid, i.e. its density, viscosity and surface tension. For the case of small amplitude oscillations, the frequency and amplitude decay rate can relatively be easily evaluated. The purpose of the experiment described here, is to apply this phenomenon as a straightforward method of measuring surface tension for gas-liquid systems.

An approximate analysis of small oscillations of a droplet was first performed by Lamb[1]. For the limit of small oscillation amplitudes and negligible viscosity effects compared with the surface tension of the liquid, the expression obtained by Lamb for the frequency of the n-th mode of oscillation is:

$$\Omega_n^2 = \frac{(n-1)\cdot(n+1)\cdot(n+2)\cdot n\cdot\sigma}{[(n+1)\cdot\rho_i + n\cdot\rho_e]\cdot R^3} \qquad (1)$$

where σ is the surface tension, $\rho_{i,e}$ density of the internal and external phase and R the radius of the droplet at equilibrium. The values of $n = 0,1$ have no significance as they describe compression and displacement motions. The dominating mode of oscillation is associated with n=2 and the above formula for a liquid droplet suspended in air will have the following simple form:

$$\Omega_2^2 = \frac{8\cdot\sigma}{\rho_i\cdot R^3} \qquad (2)$$

This equation is independent of the fluid viscosity and gives the asymptotical value of the oscillation frequency.

An analysis applicable to a viscous droplet oscillating in a vacuum has been first performed by Chandrasekhar[2,3] and Reid[4], later also by Valentine et al.[5]. The general solution for a viscous droplet oscillating in another viscous liquid was given by Miller and Scriven [6]. However, in the limiting case of droplets of low viscosity oscillating in vacuum or in a gas, all solutions are considerably reduced and the oscillation frequency becomes equivalent to that given by Lamb for the nonviscous case (eq.1). In the presence of viscosity,

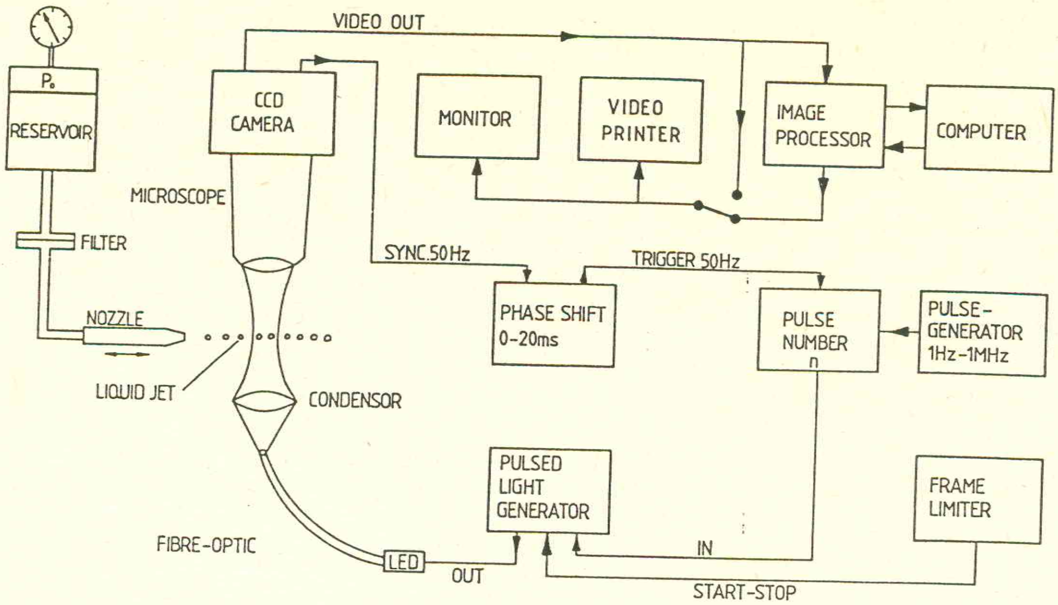

Fig.1. Experimental set up.

the oscillations are damped so that their amplitude A will decrease exponentially:

$$A_n = A \cdot \exp(-t/\tau_n) \tag{3}$$

In the limit of low viscous liquids the amplitude decay factor τ_n of n-th mode of oscillation obeys the following equation:

$$\tau_n = \frac{R^2}{\nu \cdot (n-1) \cdot (2n+1)} , \tag{4}$$

where ν is the kinematic viscosity. For a water droplet of 0.1 mm radius we obtain for n=2 $t_2 = 2 \cdot 10^{-3}$s and for n=3 $t_3 = 7.1 \cdot 10^{-4}$s. The third mode is damped almost three times faster than the second one.

The validity of the model of periodical oscillations is limited by the viscosity of the medium and radius of the droplet. The theoretical considerations of this problem by Chandrasekhar[3] and Reid[4] determined the manner by which the excited droplet returns to the equilibrium state. With the help of the characteristic parameter α, defined as

$$\alpha_n^2 = \frac{\Omega_n \cdot R^2}{\nu} , \tag{5}$$

they could show that for values of α^2 which exceed a certain critical value, damped oscillations will occur, while for values of α^2 smaller than this critical value, two aperiodic modes of decay appear. For the principal mode n=2 this critical value is $\alpha_2^2 = 3.6902$ and for n=3 for $\alpha_3^2 = 6.026$.

For the mode n=2, which is the most interesting for our purposes and in the case of small viscosity we find by applying (2) for the oscillation frequency, that the critical radius of the droplet is equal to:

$$R_c = \frac{1.702 \cdot \rho_l \cdot \nu^2}{\sigma} \tag{6}$$

This gives for a droplet of water surrounded by air a critical radius $R_c = 2.3 \cdot 10^{-6}$cm. A deformed droplet larger than this critical radius will therefore tend to oscillate while a smaller droplet will aperiodically return to its former spherical form.

Experimental

Preliminary experiments were performed with water and alcohol droplets dispersed in air. A scheme of the experimental setup is shown in Fig.1. A pressurized tank supplies the liquid to a stainless steel nozzle that has an inner diameter of 0.1 mm. A laminar jet of a few centimeter of length and uniform diameter issues from the nozzle and due to capillary instability breaks up into a train of droplets. This disintegration process can be controlled by superimposing external disturbances to the jet velocity in order to obtain almost monodispersed droplets[7]. However, as monodispersity is not so important in the present experiment it is sufficient to only control the locus of break up of the jet by adjusting the pressure in the supplying tank. The droplets generated are of 0.2 mm to 0.4 mm diameter and move at velocities of approximately 1 m/s.

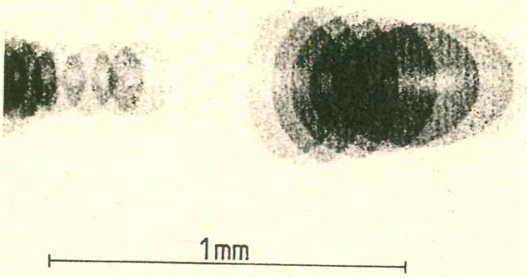

Fig.2. Multi-exposed video frame of an oscillating droplet. Strobe frequency 10 kHz, flash duration 200 ns.

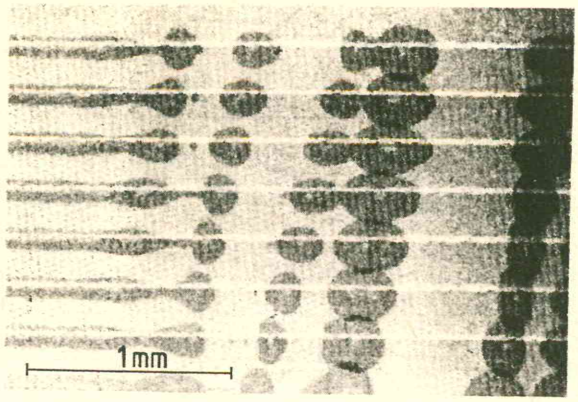

Fig.3. Electronically shifted multi-exposed video frame of jet break up. Strobe frequency 20 kHz, flash duration 200 ns.

The oscillations are observed by a video camera mounted on a microscope. A light-emitting-diode (LED), at an emission wave length of 660nm, type H 3000 (Stanley), is used for illuminating of the jet stroboscopically. The light is coupled from the diode by an optic mono-fibre to get a more homogeneous intensity distribution in the field of observation. Using a condenser lens the light beam leaving the fibre is adapted to the aperture of the microscope in order to illuminate the whole field of observation at minimum light losses. The LED is driven by a specially designed pulse generator[8] which allows to produce light flashes with a repetition frequency up to 1 MHz and a duty cycle ranging from 0.05μs to 10μs. To minimize image blurring of the quickly moving droplets the duration of light flashes should be sufficiently short. For a droplet velocity of 2 m/s and a 10-fold magnification, the maximum exposure time with respect to the spatial resolution of the camera should not exceed 1 μs. The pulse width applied in our experiments is in the range of 100-300 ns.

The image sensor of the camera is a solid-state charge coupled device (CCD) of type NAX 1011 (Valvo), of 288 (vertical) by 604 (horizontal) pixels for each of the two interlaced fields. The camera is directly coupled with an 8-bit videoprocessor which allows to store in real time 8 digitized images. Then, these images are stored by a computer on a disk for subsequent analysis.

The visualization method used in our experiment[9] is based on the principle of multiple exposure of a transient object on a single video frame. In the case discussed here we apply bright field illumination and assume that the droplets which are projected on the CCD sensor become visible as dark regions. Without any further knowledge about the nature of the object being observed one could only state how many times each pixel of the sensor has been exposed. Fig.2 gives a video print of such a multiexposed frame.

At the moment however, when we know or may assume additional details from other findings an interpretation of the picture becomes very often possible. For the case of our droplets we assume that the particles during the period of observation neither coalesce nor split up. Then, the shape of the projection of the particle at the different moments of exposure becomes immediately visible. Partly, this is also due to the fact that the object is moving quite rapidly across the field of observation so that the overlapping regions of the single superimposed images are quite restricted.

Another way to differentiate between the single images on the same frame is based on a "peculiarity" of the CCD camera. Even if the object observed performs no translational motion the video frame as a whole can be shifted electronically relative to the image of the object. This occurs during the image transfer process from the image section of the sensor to its storage section. This process takes in our case about 0.5 ms and can be prolonged. If, during this time the object is illuminated stroboscopically, the multiple exposed frame displays the phases of the droplet oscillation as a row of vertical beads (during the transfer process the whole frame moves vertically i.e. along the columns of the pixels). This is illustrated in Fig.3. Of course, also in this case overlapping image regions may occur.

In both cases, the spatial resolution is additionally connected with the possibility to detect small intensity differences on the video frame. This depends on the quality of illumination and on the dynamic range of the camera and videoprocessor. In our case, for an 8 bit processor, more than 100 grey levels of the video signal are detectable. Practically, for such a regular object as a droplet, only 10 to 20 grey levels are often sufficient to analyze a series of 20 to 30 superimposed pictures on one frame. The digital images of oscillating droplets stored in the computer memory can be either printed immediately by a video printer or first processed in the computer. The image processing software available allows by subtracting the background, gamma correction and edge preserving filters to differentiate between boundaries of overlapping droplet images indistinguishable on the real-time images. Moreover, there exist rules on the topological properties of superimposed regions which may be helpful for the coordination of intersecting boundaries of different cross sections. The so processed digital images are used subsequently to analyze the shape of the droplet.

For small amplitude oscillations the droplet preserves a spheroidal form. If the symmetry axis of the spheroid is in the plane of observation then both of its principal axes coincide with the axes of the ellipse observed on the video frame and can be directly measured. This case is quite often encountered in the experiment, as the forces acting on the jet during the break up process are axial symmetric and the direction of observation is perpendicular to the jet axis. Measurements of the ellipse principal axes can be done either directly or by a polygon (ellipse) fitting procedure, which would reduce the measurement errors.

The oscillation of the spheroid appears as a change of length of its both axes. The length of the symmetry axis C(t) of the spheroid changes in time according to the following equation:

$$C(t) = R\cdot(1 + A\cdot\sin(\Omega\cdot t + \phi) \tag{7}$$

where Ω is the angular oscillation frequency and ϕ the phase angle. As the volume V of the droplet does not change during observation time, the length of a second principal axis B(t) of the spheroid can be described for A << R by the following equation:

$$B(t) = \sqrt{\frac{3\cdot V}{4\cdot\pi\cdot C(t)}} \sim R\cdot(1-\frac{A}{2}\cdot\sin(\Omega\cdot t + \phi)) \tag{8}$$

To guarantee negligible changes of frequency Ω and amplitude A during observation, the time of registration is limited to one or maximum two periods. This fact excludes FFT methods to analyze the oscillations. Hence, the oscillation frequency is evaluated by fitting the theoretical expressions (7) and (8) to the measured lengths of both axes of the spheroid. It is realized by a computer minimization of the four parameter function

$$F(\Omega,\phi,A,R) = \sum_i [C_i - C(t_i)]^2 + \sum_i [(B_i - B(t_i)]^2 , \tag{9}$$

where Σ is the sum over lengths C_i and B_i of the axes measured simultaneously at times $t_i = i*\delta t$, with δt being the time interval between flashes.

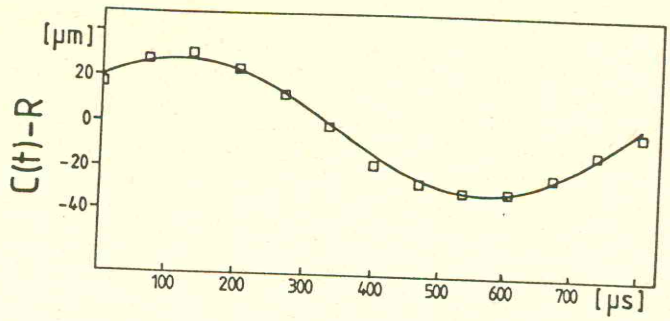

Fig.4. Example of evaluation for an oscillating ethyl alcohol droplet. Solid line - "best fit" by eq.(9), squares - measured values of lengths of axis. Results: Ω = 7.03 kHz, ϕ = 3.92, A = $3.05\cdot10^{-2}$mm, R = 0.156 mm.

First, the evaluation program checks to what extent the calculated volume of the spheroid for each pair of measured axes is constant. In the case of small deviation the starting value of radius is then evaluated from the calculated mean volume. Next, the remaining starting values for the minimization procedure are estimated. Then the program minimizes either simultaneously for both measured axes the equation (9) or, for the case that the axis of symmetry is not in the observation plane, minimizes only the first sum of (9) for each axis separately. The "best fitting" result is then taken to calculate the surface tension. Fig.4 shows an example of calculated and measured values of the symmetry axis for an oscillating ethyl alcohol droplet. The calculated surface tension, from (2) for the "best fitting" frequency Ω and radius R, is equal 18.5 ±1.0 g/cm.

Results

Preliminary experiments were performed with water and ethyl alcohol jets. The frequency of the strobe illumination was chosen in the range from 10 to 20 kHz, which gives about 10 to 20 pictures for a full period of oscillation. Among the pictures taken of the droplets we have selected only those, where undoubtedly the axis of symmetry of the droplet was in the plane of observation. The digital images registered were processed to enhance the visibility of the edges and then printed out. A provisional analysis of the oscillation used here was performed directly from the video prints by measuring the lengths of the two axes of the droplet image. Then, these sets of values were processed with the help of the above described minimization procedure to evaluate frequency Ω and droplet radius R. From these values the surface tension of the liquid was calculated using eq. (2).

A few examples of surface tension values measured in this way are collected in Table 1 and compared with data available in literature. The relative error is about 15%.

The main source of the error is due to an uncertainty in estimating the droplet radius. We hope that further progress in computer aided shape analysis will result in an apparent increase of measurement accuracy. By additional development of the evaluation procedure, it is possible to analyze images of droplets where the principal axes are not laying in the observation plane and also to employ more sophisticated, higher order oscillation models.

Nr	liquid	Strobe freq. kHz	Osc. freq. Ω kHz	Droplet radius R mm	Calc. σ g/s^2	Tab.[10] σ g/s^2
1	ethyl	15	7.03	0.156	18.5	22.39
2	alco-	15	7.12	0.151	17.2	
3	hol	15	7.19	0.150	17.3	
4	water	10	7.97	0.201	64.5	72.88
5		10	7.46	0.207	61.7	
6		20	7.65	0.204	62.1	

Table 1. Examples of measured values of surface tension.

Discussion

It appears that method proposed can be developed as a very useful tool for measuring surface tension, especially for all transient processes, where the experimental conditions are changing very quickly. As a first application of this method it is planned to study the evaporation of a single droplet moving in a vapor. A detailed description of this process is essential for understanding the heat and mass transfer mechanisms in a variety of industrial processes and applications and also in atmospheric sciences. There are two important but very difficult to measure parameters in the analysis of the evaporation of a single droplet: its surface temperature and its surface tension. (This second parameter becomes very important in the case of liquid droplets evaporating into vacuum, when the inner pressure becomes comparable with vapor pressure). As the surface tension depends strongly on the temperature described here method of measuring surface tension may also, by proper calibration, offer an information about the surface temperature of the evaporating droplet.

Moreover, this method combining computer image processing with CCD camera high speed video registration, can also be applied in several other experiments, saving time of tedious analysis of high speed 16 mm movie pictures. However, the method is limited to objects with well defined edges and where registration of only 20-30 images of the transient state provides sufficient information. Hence, it is particularly useful for observation of such processes like oscillations and unsteady motion of mechanical objects i.e. bubbles and particles, propagation of cracks in solid bodies, cavitation, boiling, and evaporation of droplets.

Literature

1. Lamb H., (1932). Hydrodynamics, sixth edition. Cambridge University Press, Cambridge. pp. 473-475.

2. Chandrasekhar S. (1959) The Oscillations of a Viscous Liquid Globe, PROC. LONDON MATH. SOC., 9: pp.141-149.

3. Chandrasekhar S. (1961). Hydrodynamic and Hydromagnetic Stability. Clarendon Press, Oxford. pp.466-477.

4. Reid W.H. (1960). The Oscillations of a Viscous Liquid Drop. QUART. APPL. MATH. 18: pp.86-89.

5. Valentine R.S., Sather N.F., Heideger W.J. (1965). The Motion of Drops in Viscous Media. CHEM. ENG. SCI. 20: pp.719-728.

6. Miller C.A., Scriven L.E. (1968). The Oscillation of a Fluid Droplet Immersed in Another Fluid. J.FLUID MECH. 32: pp.417-435.

7. Chaudhary K.C., Maxworthy T. (1980). The Nonlinear Capillary Instability of a Liquid Jet. Part 3. Experiments on Satellite Drop Formation and Control". J. FLUID MECH. 96: pp.287-297.

8. Hiller W., Lent H.-M., Meier G.E.A., Stasicki B. (1987). A Pulsed Light Generator for High Speed Photography. EXP. IN FLUIDS. 5: pp.141-144.

9. Hiller W., Kowalewski T.A. (1987). Eine Einfache Hochgeschwindigkeitskamera mit CCD-Sensor. BERICHT NR.8/1987, Max-Planck-Institut für Strömungsforschung, Göttingen.

10. Jasper J.J. (1972). Surface Tension of Pure Liquid Compounds. J. OF PHYS.CHEM. REF. DATA. 1: pp.852,948.

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