Review of Scientific Instruments

A new method for measuring liquid surface tension with acoustic levitation

Cite as: Review of Scientific Instruments **66**, 3349 (1995); https://doi.org/10.1063/1.1145506
Submitted: 01 December 1994 . Accepted: 03 January 1995 . Published Online: 04 June 1998

Yuren Tian, R. Glynn Holt, and Robert E. Apfel

A new method for measuring liquid surface tension with acoustic levitation

Yuren Tian, R. Glynn Holt, and Robert E. Apfel
Department of Mechanical Engineering, Yale University, New Haven, Connecticut 06520

(Received 1 December 1994; accepted for publication 3 January 1995)

A new method utilizing an acoustic levitation technique is introduced to measure liquid surface tension. During the measurement a small drop of test liquid is acoustically levitated in air, and its static shape is gauged with the variation of its altitude location. The experimental data are matched with theoretical calculations giving an estimation of liquid surface tension. Due to its noncontact manipulation and requirement for a small amount of test sample, this method can be applied in many special situations. © *1995 American Institute of Physics.*

I. INTRODUCTION

Surface tension can be measured by many different techniques.[1] For most conventional methods, the tested liquid sample has to be in contact with a solid material such as a ring, plate, or capillary. Therefore, during the measurement one must carefully clean the experimental hardware and use caution to prevent contamination from the environment. Further, according to the wetting situation, different kinds of solid materials may be used for different liquids, and the calibration of the system can take much effort.

Acoustic levitation provides a noninvasive method to measure the surface tension. During the test a small drop of the liquid is levitated and isolated within another medium (liquid or gas) by an acoustic radiation force. Usually the levitated drop is excited into shape oscillations, and the surface tension is determined by measuring the resonance frequency.[2–4] However, in ground-based (1 g) experiments, the levitated drop has a nonspherical static shape due to the gravitational force and the acoustic radiation stress on the droplet surface. Since the theoretical analysis of the shape oscillation of a deformed drop involves many complexities, the interpretation of experimental data becomes difficult. In particular, the inference of the static surface tension from frequency data is complicated by frequency shifts because of droplet deformation.[5,6] On the other hand, since the static shape of a levitated drop is not only a function of gravitational force and acoustic radiation stress, but also depends on the droplet surface tension, it is possible to evaluate the liquid surface tension by gauging the droplet static shape in a levitating sound field. In this paper we will present a new method to measure the liquid surface tension by detecting variations of droplet aspect ratio versus the sound pressure or, equivalently, the droplet altitude position. Unlike the technique of droplet shape oscillation, the method introduced here is designed to measure the liquid static surface tension. However, for many liquids, such as surfactant solutions, their static surface tensions can be different from the dynamic ones. Therefore, the shape oscillation method is still desired for these cases.

II. THEORY OF DROPLET DEFORMATION IN A LEVITATION SOUND FIELD

The deformation of an acoustically levitated liquid drop in a sound field has been theoretically analyzed in a previous paper.[7] If the drop is symmetric about its central axis, which follows the direction of gravity, and has an equilibrium oblate shape, the droplet profile can be described by (see Fig. 1)

$$r_s(\theta)=R[1+f(\theta)], \tag{1a}$$

where

$$f(\theta)=\sum_{l=0}^{\infty} A_l P_l(\cos\theta). \tag{1b}$$

For this axisymmetric representation of droplet static deformation, R is the droplet radius in the absence of the sound field and gravitational force, $P_l(\cos\theta)$ is the lth order Legendre function, and the A_l are constant coefficients.

In principle, the droplet surface curvature is determined by solving the Young–Laplace equation,

$$\Delta P=\gamma\nabla\cdot\mathbf{n}, \tag{2}$$

where ΔP is the pressure difference across the droplet surface, γ is the surface tension, $\mathbf{n}$ is the outwardly directed unit normal of the surface, and $\nabla\cdot\mathbf{n}$ is the total surface curvature.

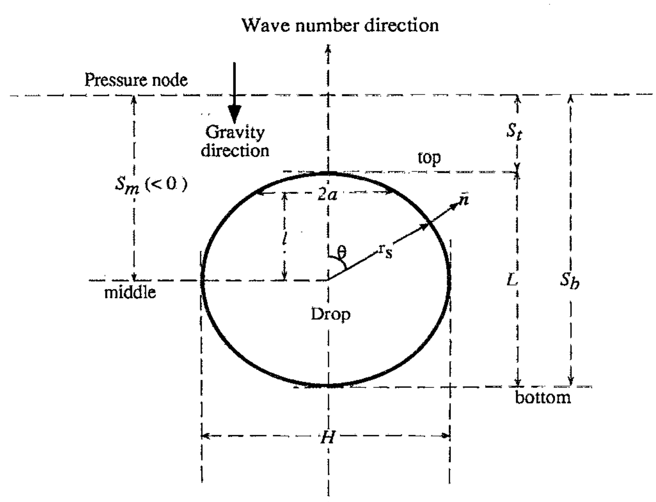


FIG. 1. Schematic description for the sideview of an acoustically levitated liquid drop in air. S_t, S_m, and S_b are, respectively, the locations of top, middle, and bottom points of drop, θ is polar angle, $2a(l)$ is the hypotenuse length, H is the diameter of drop equator, and L is the length of droplet center axis.

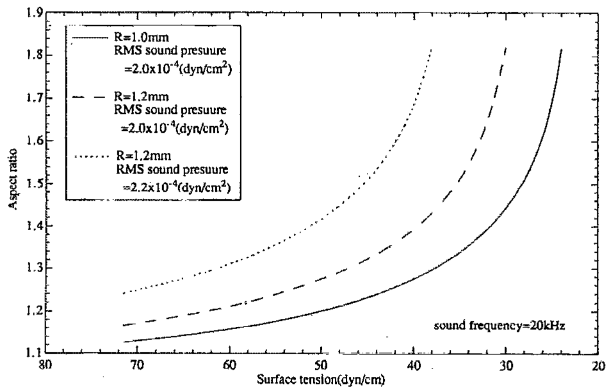


FIG. 2. Theoretical calculations of aspect ratio vs surface tension of liquid drops acoustically levitated in air for given sound pressure and droplet volume in 1 g. The density of liquid is taken as 1 g/cm^3, and that of air as 1.22×10^{-3} g/cm^3. The sound speed in the liquid is 1498 m/s, and 340 m/s in air. The sound frequency is 20 kHz.

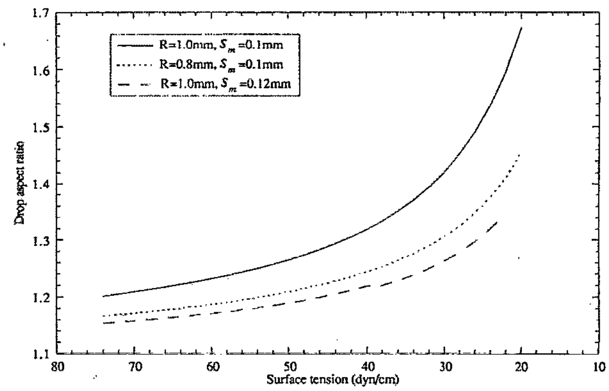


FIG. 3. Theoretical calculations of aspect ratio vs surface tension of liquid drops acoustically levitated in air for given droplet altitude and droplet volume in 1 g. The levitation sound frequency =27.9 kHz.

When a liquid drop is acoustically levitated in a gravitational environment, ΔP can be divided into three components;

$$\Delta P=\Delta P_{\rm rad}+\Delta P_g+\Delta P_{\rm st}. \tag{3}$$

Here, $\Delta P_{\rm rad}$ is the contribution from the acoustic radiation, ΔP_g is induced by gravity, and $\Delta P_{\rm st}$ is the difference of uniform static pressures inside and outside of the drop. In Ref. 7, the theoretical expressions for $\Delta P_{\rm rad}$, ΔP_g, and $\Delta P_{\rm st}$ have been derived, and a numerical algorithm has been developed to calculate the droplet surface shape and position under a given sound pressure, droplet size, and surface tension. Since the multiple interactions between the drop and sound field, the acoustic scattering by a nonspherical object and the limitation of droplet volume variation during the shape change have been considered in the theoretical analysis. This theory is valid for drops with aspect ratio as large as 2.

In order to evaluate the liquid surface tension by gauging the droplet static shape, the relation between the droplet aspect ratio and the liquid surface tension should be determined. Figure 2 plots the droplet aspect ratio versus liquid surface tension for a given droplet size and sound pressure. It shows that the droplet aspect ratio is a sensitive function of liquid surface tension, especially when the liquid surface tension has a small value. During the experiment, three parameters—droplet size (or volume), aspect ratio, and sound pressure—need to be measured. Usually the measurement of sound pressure is complicated and the accuracy is limited. However, with the variation of sound pressure, the equilibrium levitation position of the drop will also change. Since the sound pressure and the droplet altitude position have a one to one relationship, the measurement of sound pressure can be replaced by the measurement of droplet location which can be accomplished very easily and accurately. Figure 3 displays the relationships between the droplet aspect ratio, altitude location, and the liquid surface tension. Thus, the droplet surface tension can be inferred by the following two steps: first the relationship between droplet aspect ratio and altitude location is experimentally determined; then the liquid surface tension is evaluated by fitting the experimental data with the theoretical calculations, using surface tension as an adjustable parameter.

III. APPARATUS

The experimental apparatus is composed of two systems: an acoustic system and an observation system (see Fig. 4).

The acoustic system is similar to one used in a previous work[8] which employs a horn-shaped "sandwich" transducer, a focusing reflector, and a test chamber. The transducer consists of two aluminum hollow cylinders, one aluminum horn, and two 2-in.-diam, 1/8-in.-thick PZT (piezoelectric lead–titanate–zirconate) hollow disks (Channel Industries, Inc., CA). All of these elements are held together and prestressed by a central bolt. The resonance frequency of this transducer is 28.2 kHz and the Q value is around 100. The two PZT disks are excited into vibrations with opposite direction so that a velocity node is located at the positive electric terminal. The total length of the transducer is about one wavelength. In order to effectively generate uniform plane waves in air, the diameter of the horn tip should be larger than the sound wavelength in air. A 1.75-in.-diam, 1/8-in.-thick aluminum disk is attached to the tip of the horn so that the radiation area is increased.

The design of the reflector has to meet two requirements: it should provide a method to detect the sound pressure during the measurement, and the observation of the liquid drop from the top should be possible. A small glass window is built at the apex of the reflector. Between the glass plate and the shell of the reflector there is a poly-vinylidene fluoride film (PVDF from Atochem North America, Inc., PA) which works as an acoustic probe. A small hole is cut at the center of the PVDF probe thereby permitting the observation of the test sample from the top. When the sound pressure at the surface of the reflector reaches 120 dB, the output signal from PVDF probe is larger than 10 mV.

Both the acoustic transducer and reflector are placed inside a test chamber. When a resonant standing wave along the direction of gravity is excited in the air gap, a liquid drop can be levitated in air. The test chamber is carefully sealed and can be filled by a desired gas such as pure nitrogen. At

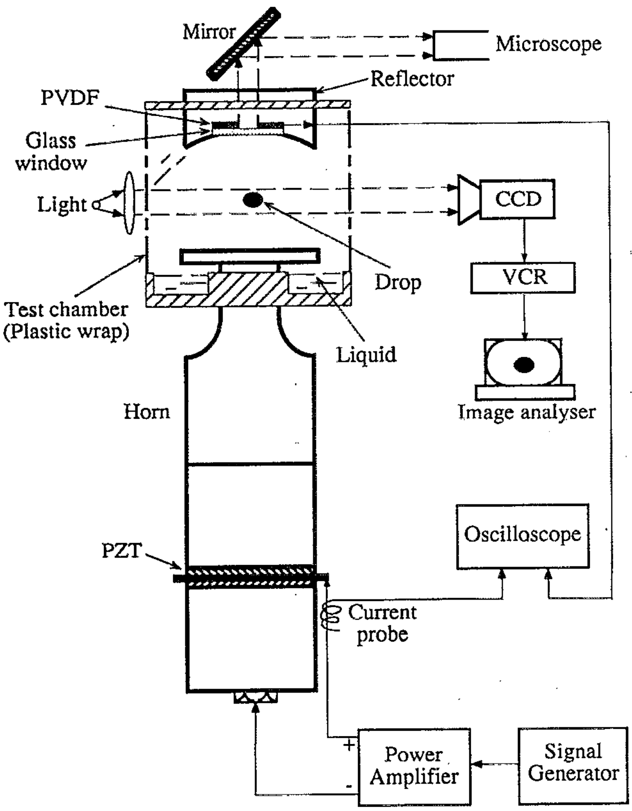


FIG. 4. Diagram of the experimental apparatus.

the bottom of the chamber there is a trough. When it is filled with the liquid, the evaporation of the levitated drop can be reduced. The acoustic boundary condition at the side of the test chamber is very important to the control of the experiment. A pressure-release boundary condition reduces the unexpected distortion of the symmetry of the sound field inside the chamber, and maintains the levitated drop in a closely symmetric static shape. As shown in Fig. 4, the test chamber is built without a rigid sidewall and is covered with a thin sheet of plastic wrap. Hence the acoustic wave can propagate from inside to outside.

The whole acoustic system is seated on a vibration-isolated table. The alignment between the transducer and reflector is adjusted by micropositioners. Two tilt/rotation stages are used to keep the surfaces of the transducer and reflector parallel to sea level.

The static shape of a levitated drop is observed through a microscope and a high performance CCD camera (mode 4915, Cohu, Inc., CA). An optical mirror mounted above the acoustic reflector is used to obtain the image of the test sample from the top, and the droplet side image is recorded by the CCD camera and saved on a video tape. The application of diffusive background lighting produces a dark picture of the drop with a very clear edge. During the replay of the tape, the droplet side image is digitized and analyzed by an image analysis system (Acuity, Inc., MA). The droplet edge is searched along horizontal lines and following the direction from the outside to the inside of the drop. Both variations of the intensity and intensity gradient are used to determine the locations of droplet edge points; usually the uncertainty is

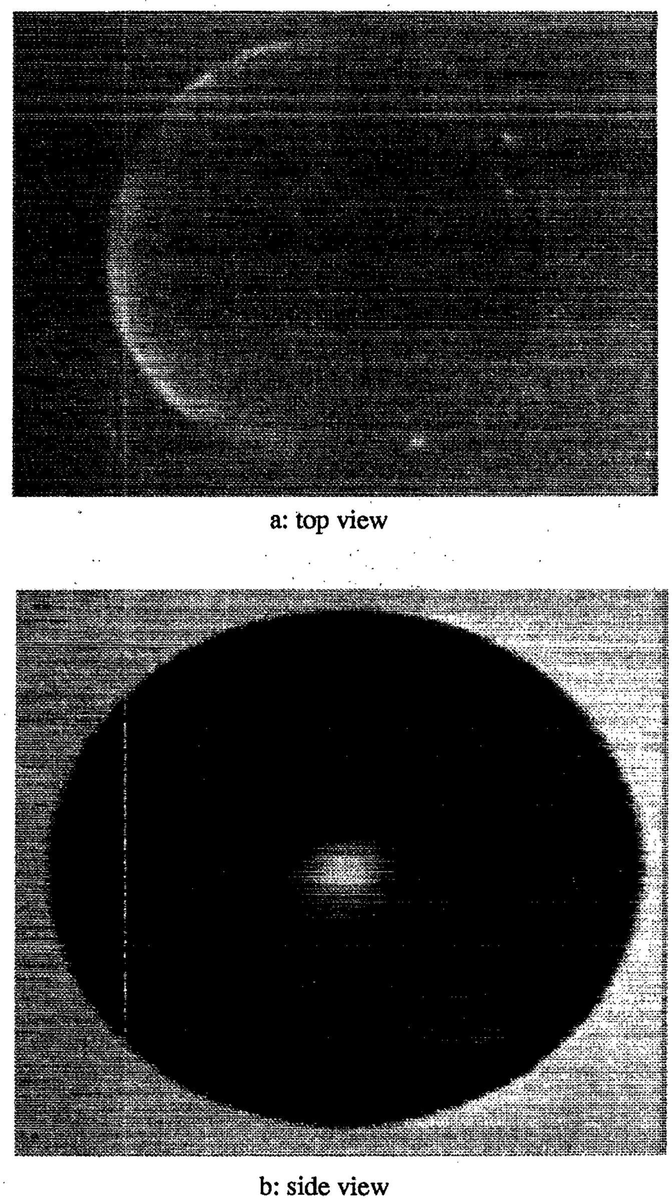


FIG. 5. Video images of the static shape of a levitated water drop; (a) Top view, (b) sideview. R=1.1 mm. Frequency of the levitating sound =28.1 kHz.

less than 2 pixels. The whole image system is calibrated with a micrometer (1/100 mm Divs., Edmund Scientific), and the resolution on a monitor of 640×480 pixels is better than 6.3 μm/pixel.

IV. MEASUREMENT RESULTS AND DISCUSSIONS

The liquid drop can be manually injected inside the test chamber through a small window by a glass syringe and a hypodermic needle. After the drop is levitated, the symmetry about its vertical (gravitational) axis is judged from the top view. With the image analyzing system, the sideview gives the drop's vertical and horizontal dimensions (L and H), aspect ratio (H/L), volume, and the spatial position ($S_{t,m,b}$). (See Fig. 1.)

Figure 5(a) gives one example of the droplet top view. In

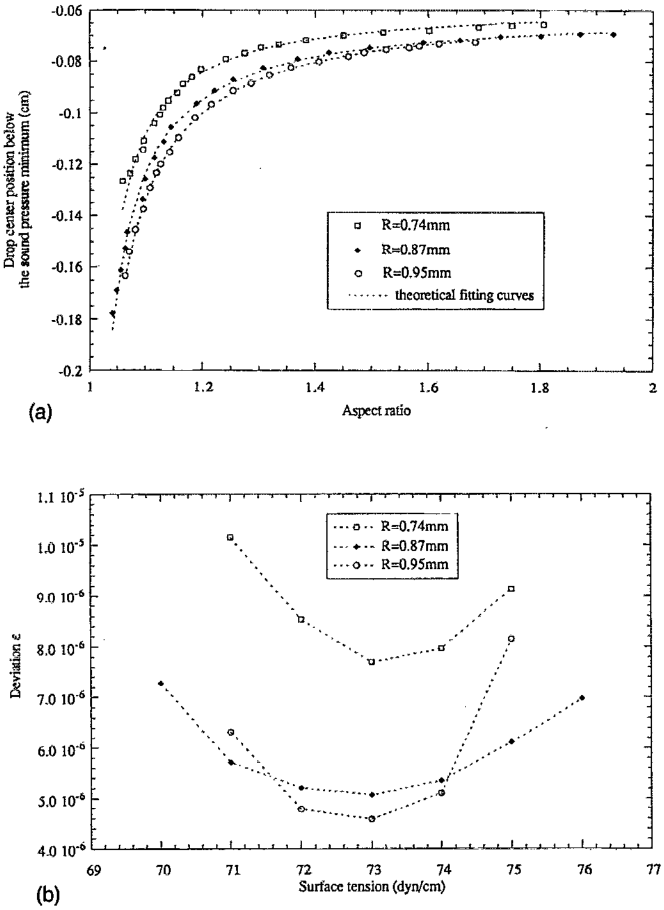


FIG. 6. Measurements of the static deformation of water drops levitated in a sound field. (a) The variation of droplet center position versus the aspect ratio. (b) The deviation between the experimental data and the theoretical matching. Frequency of the levitating sound =27.9 kHz.

our experiments the ratio of the maximum diameter to the minimum of the droplet top view is very close to one. (The deviation is less than 1%.) Therefore the levitated drop can be considered symmetric about its vertical axis. The volume of the drop may be calculated from the image of the sideview [Fig. 5(b)]. Since the drop has an equilibrium oblate shape and its profile is symmetric about the central axis, the volume can be found as

$$V=\pi\int_0^L a(l)^2 dl, \tag{4}$$

and the volume-equivalent droplet radius is

$$R=(3V/4\pi)^{1/3}, \tag{5}$$

where $2a(l)$ is the hypotenuse length. In the experiment it is determined by counting the pixel numbers along the horizontal lines within the droplet side image.

As the pressure of the levitating sound field varies, both the droplet aspect ratio and spatial altitude will change. In order to find experimentally the relationship between these two parameters, the levitated drop is squeezed into an oblate shape with an aspect ratio about 2 by using an intense levitation sound field. By gradually reducing the sound intensity, the droplet aspect ratio is decreased and its altitude is lowered until it moves out of the observation range. From the recorded images of the drop, the droplet aspect ratio and the spatial altitude $S_{t,b,m}$ can be determined. The equilibrium surface tension of the liquid is found by matching the experimental raw data with the theoretical calculations.

For each individual drop, if the total number of the experimental data points is N, the equilibrium surface tension γ_{eq} should make the value of ϵ in

$$\epsilon=\sum_N [S_{t,m,b}(A_r)_n - S^c_{t,m,b}(A_r,\gamma)_n]^2 \tag{6}$$

approach a minimum. Here $S^c_{t,m,b}(A_r,\gamma)_n$ is the theoretical calculation of droplet altitude position for a given aspect ratio A_r and surface tension γ. This is essentially a process of nonlinear least-squares fitting. However, since the theoretical treatment introduced in Ref. 7 cannot be easily used in the standard nonlinear least-squares fitting algorithm, a simplified alternative is used here. In this routine, the surface tension γ is used as an independent variable, and for its different values, the magnitude of ϵ is calculated. If the variation range of γ is properly selected, ϵ will become a minimum at one of the selected surface tensions, and the corresponding value gives the best estimation of the equilibrium surface tension of the liquid. The accuracy in this evaluation is con-

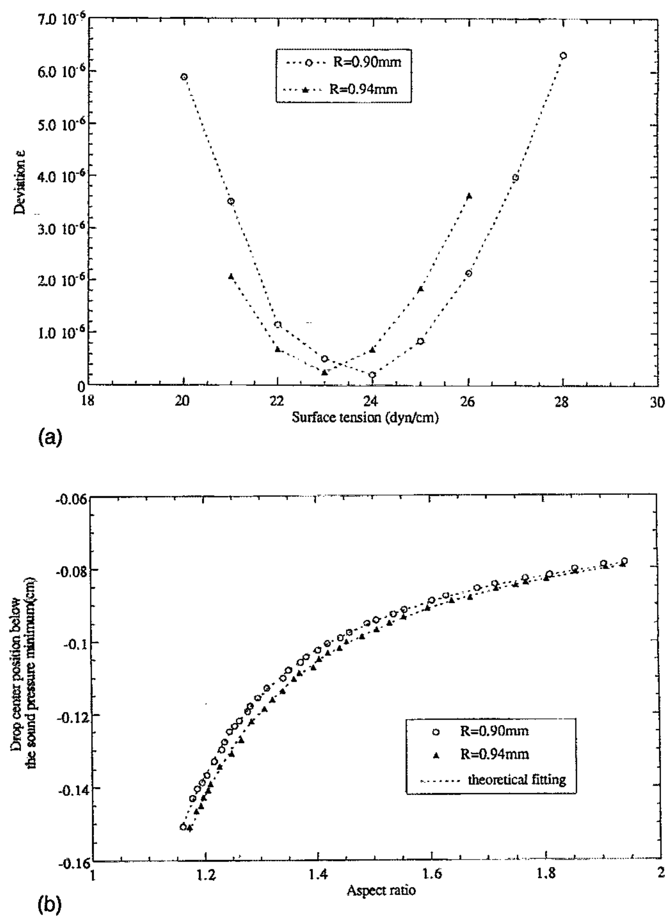


FIG. 7. Measurements of the static deformation of dodecane drops levitated in a sound field. (a) The variation of droplet center position vs the aspect ratio. (b) The deviation between the experimental data and the theoretical matching. Frequency of the levitating sound =27.9 kHz. The density of dodecane is taken as 0.78 g/cm^3.

TABLE I. Experimental results of water and dodecane. Temperature =23 ±1 °C.

	Properties			
	Surface tension (dyn/cm)			
Materials	Drop deformation	Ring	Reference[a]	Density[b] (g/cm^3)
Water	73±1	72.3	72.9 (20 °C)	0.997 (20 °C)
Dodecane	23.5±1	23.8	25.3 (20 °C)	0.748

[a]Reference 13.
[b]Reference 14.

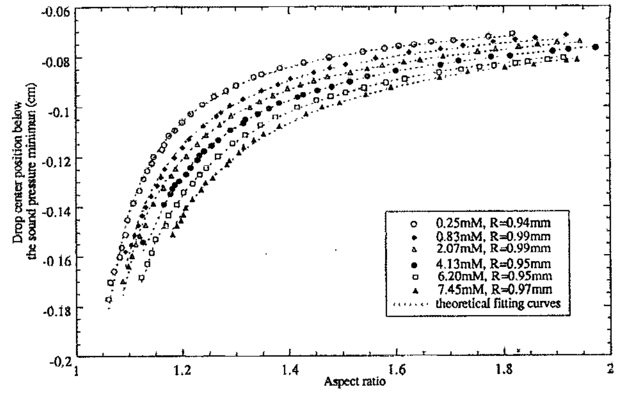
trolled by the step size of γ when it varies. In the present studies, the step size is always taken as 1 dyn/cm.

Our technique is tested by measuring the surface tensions of pure water and dodecane. The measurements are performed at the room temperature, which is maintained around 23±1 °C. The experimental raw data with the theoretical matching curves are presented in Figs. 6 and 7, and the magnitude of ϵ under different values of γ are also displayed in Figs. 6 and 7. From these plots the surface tension of pure water can be estimated as 73±1 and 23.5±1 dyn/cm for dodecane. The surface tensions of water and dodecane used in our tests are also measured with the ring method (Fisher surface tensiometer, model 20). Table I compares our measurement results obtained by two different methods with some literature values and shows that the new technique introduced in this paper can give a reliable measurement of the liquid equilibrium surface tension.

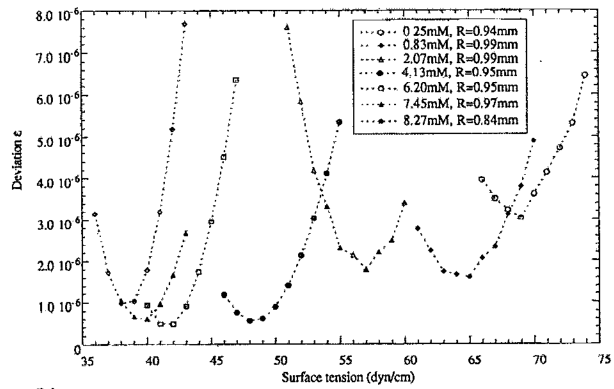
Some surfactant solutions are also tested with our systems. Figure 8 gives the measurement results of the equilibrium surface tensions of sodium dodecyl sulfate (SDS) aqueous solutions under different concentrations. The surface tension of SDS aqueous solution has also been studied by many other authors with different methods[9–11] and their measurement results agree fairly well with ours (Fig. 9).[12]

Drops of different sizes can be used in the measurement. For our apparatus, drops with radius between 0.8 and 1.1 mm are the best choice for the measurement. A larger drop can easily become unstable and break up; for a smaller one, its altitude displacement with the change of the aspect ratio becomes so small that the quality of the matching between the experimental data and the theoretical fitting curve is reduced. In order to achieve high sensitivity for a liquid with high surface tension, a large drop should be used. From Fig. 3, it can be seen that if the precision of the measurement of droplet aspect ratio is 1%, the sensitivity in the measurement of surface tension is better than ±2 dyn/cm when the surface tension is less than 50 dyn/cm. Since, in the experiments, more than one data point is used, it can be expected that the total sensitivity of our system is actually better than ±1 dyn/cm. Thus, it is plausible to assume that a smaller step size in γ could yield a smaller variance in the inferred surface tension than reported here.

The apparatus arrangement can be further improved for a practical application. In fact, during the experiment it is unnecessary to obtain the whole image of the drop because only the aspect ratio and altitude position of the tested drop



(a)



(b)

FIG. 8. Measurements of the static deformation of SDS drops levitated in a sound field. (a) The variation of droplet center position vs the aspect ratio. (b) The deviation between the experimental data and the theoretical matching. Frequency of the levitating sound =27.9 kHz. The density of SDS aqueous solutions is taken as 1 g/cm^3. (mM: micromolarity.)

are required for the measurement of the surface tension. Therefore, the image analysis system could be replaced by a simple optical detector making the whole device much more compact.

On the other hand, if the aim was to increase accuracy, the crude aspect ratio measurement could be replaced by fitting of the detected drop edge with a Legendre function

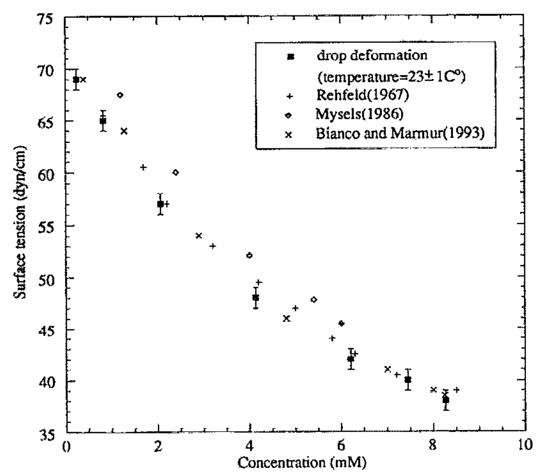


FIG. 9. Surface tensions of SDS aqueous solutions vs the concentration.

expansion. The experimental coefficient A_l at like order could then match with their theoretical counterparts, producing a more sensitive (and also more time-consuming) estimate of the surface tension.

ACKNOWLEDGMENT

The research described in this paper is financially supported by NASA through JPL Contract No. 958722.

[1] A. W. Adamson, *Physical Chemistry of Surfaces,* 5th ed. (Wiley, New York, 1990).

[2] P. L. Marston and R. E. Apfel, J. Acoust. Soc. Am. **67**, 27 (1980).

[3] C. J. Hsu and R. E. Apfel, J. Colloid Interface Sci. **107**, 467 (1985).

[4] E. H. Trinh, P. L. Marston, and J. L. Robey, J. Colloid Interface Sci. **124**, 95 (1988).

[5] E. H. Trinh, A. Zwern, and T. G. Wang, J. Fluid Mech. **115**, 453 (1982).

[6] P. V. R. Suryanarayana and Y. Bayazitoglu, Phys. Fluids A **3**, 967 (1991).

[7] Y. Tian, R. G. Holt, and R. E. Apfel, J. Acoust. Soc. Am. **93**, 3096 (1980).

[8] E. H. Trinh, Rev. Sci. Instrum. **56**, 2059 (1985).

[9] S. J. Rehfeld, J. Phys. Chem. **71**, 738 (1967).

[10] K. J. Mysels, Langmuir **2**, 423 (1986).

[11] H. Bianco and A. Marmur, J. Colloid Interface Sci. **158**, 295 (1993).

[12] It is important to note that the relaxation of surface tension for solutions of surfactants takes place in a finite time. The relaxation time depends upon both the bulk diffusion mass transport, and the sorption transport to and from the surface. Thus the surface tension of a levitated drop becomes a function of the time since the creation of a fresh surface (droplet formation/injection). All our measurements for SDS solutions were made 10 s after the drops were generated thus ensuring a surface thermodynamic equilibrium.

[13] S. Ross and I. D. Morrison, *Colloidal Systems and Interfaces* (Wiley, New York, 1988).

[14] *CRC Handbook of Chemistry and Physics,* 70th ed. (Chemical Rubber, Boca Raton, FL, 1990).