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CORE-CENTERING OF COMPOUND DROPS IN CAPILLARY OSCILLATIONS: OBSERVATIONS ON USML-1 EXPERIMENTS IN SPACE

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SUMMARY

Experiments on liquid shells and liquid-core compound drops were conducted using acoustic levitation, in a low-gravity environment during a Space Shuttle flight. It was observed that their inner and outer interfaces became concentric when excited into capillary oscillations. Using the existing inviscid theories, an attempt is made to explain the centering of the oscillating liquid shell. It is concluded that viscosity needs to be considered in order to provide a realistic description of the centering process.

INTRODUCTION

A compound drop[1] consists of a fluid drop enclosed inside another liquid drop. The inner drop, the outer drop and the external medium are called the core, shell, and host, respectively. When the core is a gas bubble, the compound drop is referred to as a *liquid shell*. In our studies, we are only interested in air as the host medium, and its dynamics can be neglected compared with that of the drop. In principle, any liquid can enclose any other liquid. But in practice, the compound drop system prefers to have the liquid with a lower surface tension on the outside because this configuration carries less total surface energy[2], e.g., it is much easier to enclose a water drop inside an oil drop than vice versa. The opposite arrangement is metastable: as long as the inner and outer interfaces do not touch it is neutrally stable. However, as soon as the two interfaces touch, the liquid inside will prefer to move completely or partially to the outer surface such that the system can adopt a lower total surface energy.

Like a compound pendulum, a compound drop exhibits two types of oscillations due to the surface tensions on its inner and outer interfaces: (1) the lower frequency slosh mode in which the two interfaces move opposite to each other and, (2) the higher frequency bubble mode in which they move in the same direction as each other[3]. In the idealized situation of a thin shell, in the slosh mode, the shell liquid mostly shuffles back and forth along the interfaces, whereas in the bubble mode, the shell liquid mostly moves in and out normally to the interfaces. For a liquid shell, neglecting the dynamics of air, the motion of the drop is essentially the motion of the shell liquid only. But for a liquid-core compound drop,

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the densities of the shell and core fluids are comparable such that the two motions are equally important. Moreover, for a low-viscosity liquid, with its two stress-free interfaces, the oscillation of a liquid shell can be considered as inviscid to a first approximation. But for a liquid-core compound drop, the shear between the shell and core liquids makes it unrealistic to neglect their viscosities.

In the absence of gravity, the core can sit anywhere inside the shell. When the drop oscillates, it has been observed, with no detailed documentation, that the core tends to go to the center of the drop for both the liquid shell[4] and the liquid- or solid core compound drop[5]. From a theoretical point of view, in the linear regime, the core is still neutrally stable anywhere inside the shell. Taking into account the non-linearity, the centering mechanism for an inviscid liquid shell has been studied in the thin shell limit[6] and for the finite shell thickness[7]. The two theories are formulated completely differently, but their essential results agree with each other. Their main conclusion is that in the presence of a pure mode of capillary oscillation, the bubble of the liquid shell undergoes a slow translational oscillation inside the shell, with a frequency that is proportional to the amplitude of the capillary oscillation. It should be emphasized that an oscillation about the center of the shell is not exactly the same as centering the latter implying that the bubble goes to and settles at the center. The present study provides a close observation of the centering phenomenon for both liquid shells and liquid-core compound drops.

Acoustic levitation is used. It is known that a standing sound field, through its acoustic radiation pressure, can provide a potential well at a pressure node for levitating a small sphere[8]. It is also known that if the sound amplitude is modulated at the resonant frequency of a drop, the latter can be excited into capillary oscillations[9]. These are the main principles behind the apparatus to be used for this experiment.

I. APPARATUS, MATERIALS, AND EXPERIMENTAL PROCEDURE

The experiments were performed in the Drop Physics Module (DPM) of the United States Microgravity Laboratory-1 (USML-1) on board the Space Shuttle Columbia (STS-50) during its flight from June 25 to July 9, 1992, by, astronauts Eugene Trinh and Bonnie Dunbar.

In the DPM, the injection system for deploying the drops consists of two similar arms sticking from the middle of the two opposite vertical sides of the walls, to meet at the center of the chamber when demanded. Each arm contains the injector tube for injecting the liquids or air, as desired. The arm tips provide the mechanical means to hold a drop during the injection.

Three types of liquids were used for the experiments: a silicone oil (Dow Corning 200 series, 2 cSt.), water, and water/glycerine (72/28) mixture (Table 1). The silicone oil and the water/glycerine mixture were chosen to have about the same viscosity (2 cSt). Since the liquids have different wetting properties they are handled slightly differently.

For deploying a water shell, a needle type injector is used (Figure 1). A water drop of the desired volume is first held between the tips, then an air bubble of the desired volume is injected. The tips are finally withdrawn, suddenly, leaving the liquid shell levitated at the center of the chamber. For deploying a silicone oil water/glycerine compound drop, a flat-tip injector is used. A silicone oil column of a desired volume is sandwiched between two flat tips during injection. Then water/glycerine mixture of a desired volume is injected into the oil drop. The flat tips are similarly withdrawn suddenly, leaving the compound drop in levitation.

The sound wave is modulated in amplitude at a modulation frequency that sweeps past the n=2 bubble or slosh mode frequency of the liquid shell or compound drop, looking for a resonant response from the drop and the expected centering of its core.

II. RESULTS AND DISCUSSION

The resonances of a liquid shell are far apart such that most of the time a pure n=2 mode can be excited. But for a liquid-core compound drop, the modes are closely packed. Also, their resonance peaks are less sharp due to dissipation by viscosity such that a lot of mode overlapping and coupling occurs. Visually, the excited shape oscillation of a liquid-core compound drop by-and-large consists of more than one mode.

A. Water Shell

The deployed drop had a volume of 4.15±0.1 cc, consisting of 2 cc of water and 2.15 cc of air. The average operating temperature was 25°C. The water contained some pliolite tracers (50 μm, Goodyear Chemical Co.) for monitoring any uncontrolled rotation of the drop.

Following deployment, it was found that the levitation of the drop was not easy, with the air bubble sloshing with respect to the shell when the drop oscillated translationally in the acoustic potential well. The bubble moved opposite to the drop translation, such that the center of mass of the drop moved back and forth, in the same direction as that of the motion of the drop, rather than staying still (Figures 2 a,b,c). The levitated drop had a small deformation along the z-axis of about 1 % ((a/b-l) x 100%, where a and b are the equatorial and polar radii, respectively), and an uncontrolled rotation of about 0.15 rps. along the y-axis. The side-view was the most suitable for observation of the interface. In a liquid-air interface, which is furthermore curved, optical distortion is an issue; however no corrections have been made for this in the measurements.

The amplitude of the sound wave in the z-direction was varied at a modulation frequency, sweeping through the n=2 bubble mode oscillation resonance of the drop from 9 to 11 Hz in 200 seconds. It was observed that the bubble was centered quickly in the first eight seconds, i.e., about 60 shape oscillation cycles. When this occurs (Figures 2 d,e), the translational oscillation of the drop in the z-direction also stopped. The translations along the x- and y-axes persisted, although the shell looked centered in those directions. It is noted that after the drop was excited into oscillation, there was a slight additional average flattening due to the modulated radiation stress averaged over a capillary wave cycle, such that a becomes a' and b becomes b'. The shape oscillation amplitude 'e' during centering (e =(L'/W'-b'/a') X 100%; L' and W' are the height and width of the drop in maximum prolate shape), was very small, an average of about 1.2 %. Once the core was centered, it did not go off-center easily.

In Figure 3, L/W (L and W being the height and width of the oscillating drop) is plotted over time for certain intervals before and after initiation of shape oscillations. The first cluster of data points represents, with some scatter, the flattening before oscillation. The second cluster (points connected) represents the drop in a forced oscillation by the modulation radiation stress, giving the mean additional deformation, mainly due to the modulation stress. The third cluster represents a similar situation at the end of centering, showing a slightly higher oscillation amplitude.

In Figure 4, the percentage eccentricity ($\Delta L/L \times 100$) is plotted versus time, where ΔL is the thickness of the shell on one side of the shell (see inset of Figure 4), and L is the height of the drop, both being dependent on time as the drop oscillates. The initial oscillation of this quantity (region I, Figure 4) arises from the fact that the drop is translating oscillatorily in the acoustic potential well, predominantly along the z-axis at a frequency of about 0.76 rad/sec. As the drop is oscillated, the translational oscillations in the potential well die out (region II) and the drop is centered (region III). The damping of the amplitude of the translational oscillations, provides unambiguous evidence of centering.

During the n=2 bubble mode sweep, the shape oscillation amplitude increased as the drop approached resonance. The amplitude 'e' peaked at 12% at around a frequency of 10 Hz (fig. 2 e,f), approximately 100 seconds following the start of the oscillation.

The experiment was repeated with the same drop with an n=2 slosh mode sweep, from 1.5 to 3.5 Hz in 200 seconds. This experiment was started following the bubble mode sweep. Prior to initiating the slosh mode sweep, the shell was almost centered along the vertical z-axis, but was off-center along the y-axis. Figure 5 is the eccentricity plot ($\Delta L/L\%$ versus time; z-axis) showing the shell almost centered prior to starting the shape oscillations (region I). As the oscillation was started, at an average amplitude of about 2.4%, the core moved off-center for about 8 seconds or about 12 oscillation cycles (region II), before becoming centered again (region III). This off-centering is probably a transient readjustment resulting from the centering along the y-axis.

During the n=2 slosh sweep, as the oscillation amplitude further increased to about 11%, with the drop approaching resonance around 2.4 Hz, the core went off center, and sloshed around violently. Eventually, the bubble got lodged on one side of the shell which, being thin, disintegrated by atomization from capillary ripples forced by the acoustic radiation pressure[11-13]. The loss of centering, at large amplitudes, is not well understood at this time.

B. Liquid-Core Compound Drop

The silicone oil contained pliolite tracers, and the water/glycerine mixture contained green food coloring (0.5% by volume). The tracers were used for monitoring any uncontrolled rotation of the drop. The dye helped to make the inner interface more visible. For an n=2 slosh mode oscillation of a liquid-core compound drop (as opposed to a liquid shell), the shell liquid tends to pile up at the equator during the oblate phase of the oscillation, forming a protrusion along the equator, which is susceptible to outward stretching by the acoustic suction stress[11]. Hence in the top view, the shell size is a little magnified. The top view was most suitable for studying the dynamics of the interfaces of the compound drop.

The drop had a volume of 3.55±0.08 cc, with 1.55 cc of oil. The average temperature during the experiments was about 24.5°C. To keep the drop from uncontrollably rotating during levitation, the drop was acoustically flattened at the poles. The resulting flattening was about 20%. In addition, an acoustic torque[10] was used to cancel any residual rotation.

The modulation frequency was swept from 2.8 to 3.8 Hz in 132 seconds, through the n=2 bubble mode, which was also close to the n=4 slosh mode of the compound drop. Four sweeps were conducted with different drive amplitudes. Initially some complex mode, mostly n=4 slosh mode, with its non-axisymmetric components up to m=±4, appeared. Then the n=2 and m=0 mode appeared, followed by a strong n=2 and m=±2 component, and next by a strong n=2 and m=+1 component. It seems that not a pure mode, but a superposition of modes with a dominant one appeared at any time. The order of the appearances did not seem to be related to the oscillation amplitude. The ease at which the highest non-axisymmetric modes occur, e.g. m=±2 for n=2 and m=+4 for n=4, can be explained by the fact that they are of the longest average wavelength among their co-modes for the same n, which implies that they are the least suppressed by viscosity. Their occurrence probably has to do with some slight biases imposed by the x- and y-drives: the slight deviation of the static shape from axisymmetry leads to a similar deviation in the modulation stress felt by the drop.

From the top-view, centering was effective with the m=±2 mode, but not much centering was seen for the m=0 and m=±1 modes. It may be concluded here that in order for centering in the equatorial plane to be effective, the shell liquid has to set up an uneven average pressure distribution through Bernoulli effect in the equatorial plane, which requires the m=±2 mode. Once the core was centered, it did not go off center, even as the amplitude of oscillation increased.. Figure 6 shows the progressive stages of centering when the m=±2 mode came into effect. Figure 7 shows a complete oscillation of the centered compound drop. A plot of percentage eccentricity $\Delta W/W*100$ versus time is given in Figure 8, showing strong evidence for centering.

The experiment was repeated for the n=2 slosh mode. The modulation frequency was swept in the range 1 to 2 Hz for 132 seconds. A few sweeps were conducted with different drive amplitudes. The drop was flattened by about 30% for levitation stability, and for driving larger amplitudes. At high oscillation amplitudes, mode coupling of n=2 mode with n=2, m=±2 mode was seen. Good, centering was achieved at small and moderate amplitudes, but at extremely large amplitudes ($e \succ 100\%$) the core went off center, because the protrusion at the equator became asymmetric due to uneven stretching by the acoustic suction stress (Bernoulli suction) at the edge. The shell finally disintegrated on the thin film, following the excitation of small capillary ripples there. The thin film is probably an outcome of the loss of stability at the edge of the protrusion[11].

The decay of the slosh mode oscillation of the compound drop, of which the inner and outer liquids had the same viscosity of 2 cSt, occurred on a time scale about 10 times shorter than the corresponding simple drop with the same viscosity, implying a strong dissipation at the inner interface.

III. THEORETICAL CONSIDERATIONS

We are interested in the centering of a liquid shell, which has been a subject of theoretical studies. Lee and Wang[6] have studied the centering of a liquid shell undergoing capillary oscillation in the thin shell limit. In this limit, the dimensionless shell thickness parameter $\delta = t_s/R_s$ is assumed to be much less than 1, where t_s is the concentric shell thickness, and R_s is the mean radius of the shell ($R_s = (R_o+R_i)/2$; R_i is the inner radius R_o is the outer radius). Because of the disparity between the frequencies of the bubble mode and the slosh mode for a given shape oscillation mode (n=2 for most studies), in the thin shell limit, different scalings are applied to the two modes.

For a liquid shell of density ρ and surface tension σ, for the bubble mode of oscillation, time is scaled by the inverse of $\omega_s/\delta^{1/2}$, where $\omega_s = (\sigma/\rho R_s^3)^{1/2}$. The wave amplitude is characterized by ε_1, which is the surface displacement amplitude scaled by R_s as in the initial condition $\tilde{R} = R_s(1 + \varepsilon_1 P_n(\cos\theta$)) relative to the geometrical center of the two surfaces ($\tilde{R}$ is the position of the mid-surface between the inner and outer oscillating surfaces, P_n is a Legendre polynomial, and θ is the polar angle). The initial displacement of the core is represented by Δ in $t = t_s(1 - \Delta\cos\theta)$, where t is the thickness distribution of the shell. It was found that for a shell with an off-center gas core, the core position undergoes a fast oscillation with the same frequency as the wave, together with a slow oscillation, about the center of the drop due to the nonlinearity of the wave; the latter being interpreted as a manifestation of the Bernoulli effect. In this study, the dimensionless frequency of the slow oscillation of the core for the n=2 bubble mode is about 1.3 times ε_1.

For the slosh mode, time is scaled by the inverse of $\omega_s\delta^{1/2}$. The wave amplitude is characterized by a ε_2 (different from ε_1), which is the amplitude of the variation in the thickness of the shell scaled by t_s, as in the initial condition $t = t_s(1 - \Delta\cos\theta + \varepsilon_2 P_n(\cos\theta))$, where t is the shell thickness distribution, and Δ represents the core displacement. It was again found that for a shell with an off-center gas core, the core position undergoes a fast oscillation with the same frequency as the capillary wave, together with a slow oscillation, about the center of the drop due to the nonlinearity of the wave. The dimensionless frequency of the slow oscillation of the core for the n=2 slosh mode is about 0.6 times ε_2. For both the bubble and the slosh mode, the results do not depend on Δ except for the extreme case of Δ close to 1, and do not depend on, or are not very sensitive to, δ as long as it is small.

Pelekasis et al.[7] considered a liquid shell with a finite thickness. Their results agree with those of Lee and Wang in the thin shell limit. Pelekasis et al. do not discriminate between the bubble mode and the slosh mode in their scalings; time is scaled with $\omega_o = (\sigma/\rho R_o^3)^{1/2}$, and lengths are scaled with R_o. For both modes, the wave amplitude is characterized by ε as in the initial condition $F_2 = 1 + \varepsilon P_n(\cos\theta) + \ldots$ for the position of the outer surface. With R* being the ratio R_i/R_o, their results for the slow oscillation frequency can be written in the dimensionless form $\Omega^* = C_b(R^*)\varepsilon$ for the bubble mode and $\Omega^* = C_s(R^*)\varepsilon$ for the slosh mode, where the coefficients C_b and C_s are functions of R*.

Converting Lee and Wang's results into the scaling system of Pelekasis et al., C_b and C_s, from both works, are plotted against R* in Figure 9 and Figure 10, respectively (the data for Ω^* of Pelekasis et al. are taken from their Figure 9 (a), (b) and (c) for the bubble mode, and from their Figure 16 and their text after their equation (7.3b) for the slosh mode). From Figure 9, it is seen that C_b depends on R* but its value from Pelekasis et al. approaches the constant value of Lee and Wang asymptotically as the shell gets thin. From Figure 10, it is seen that C_s is insensitive to R*, and its value from the two works are approximately equal, considering the numerical errors involved. It is noted that the calculation of Pelekasis et al. is more prone to numerical difficulty when R* approaches 1; being unable to finish one slow cycle, such that the values of the slow frequencies there are not very accurate. With C_b and C_s given by Figure 9 and Figure 10 respectively, the slow frequency in rad/sec for the bubble mode is given by

$$\Omega = C_b(R^*)\varepsilon\left(\frac{\sigma}{\rho R_o^3}\right)^{1/2} \qquad (1)$$

and that for the slosh mode by

$$\Omega = C_s(R^*)\varepsilon\left(\frac{\sigma}{\rho R_o^3}\right)^{1/2} \tag{2}$$

where it is recalled that ε is the surface displacement amplitude scaled by R_0.

IV. COMPARISON WITH EXPERIMENTAL RESULTS

The translational oscillation in the potential well, before the liquid shell is excited into shape oscillations, has a frequency about 0.76 rad/sec (Figure 4) . After it is excited into the bubble mode, the translational frequency is about 1.6 rad/sec. The average oscillation amplitude (e %) for centering from Figure 3 is about 1.2%. For the current liquid shell $R^* \sim 0.8$ and thereby C_b is ~ 6.5 from Figure 9. Imposing this onto equation 1, while noting that ε is ~ 2/3 e, we arrive at a frequency of about 0.44 rad/sec. There is no strong evidence for the presence of this frequency. However, the time scale over which centering occurs is about 8 seconds. It is very tempting to consider this as an over damped slow oscillation, representing a one-half period, leading to a frequency of about 0.4 rad/sec. The bottom line is that the result questions the validity of inviscid assumption in the context of centering of the liquid shell.

Similarly in the context of Figure 5, for centering in the slosh-mode, the average oscillation amplitude is about 2.4 %. For the current drop, C_s is ~ 4, from Figure 10. Imposing these onto equation 2, we arrive at a frequency of about 0.54 rad/sec. Again, it is very tempting to consider the transient re-adjustment of the core (Figure 5), over a time scale of about eight seconds, as representing a half-period of slow oscillation.

CONCLUDING REMARKS

We have seen that when centering of a compound drop in capillary oscillation occurs, the core stays at the center after a few oscillations. For a liquid-core compound drop, this is reasonable because of the viscous friction between the core and shell fluids. But for a liquid shell, in the inviscid limit, it has been predicted by two independent theories that the core should go through a slow oscillation with a frequency proportional to the wave amplitude. In view of the discrepancy between the theoretical prediction and the experimental observation, the obvious question is: should the liquid shell be considered inviscid? The observation seems to suggest that as far as the nonlinear effect called centering is concerned, viscosity should be taken into account in order to obtain a realistic prediction.

We have also observed that the liquid shell can become decentered at large wave amplitude, for the case of the n=2 slosh mode. This effect could be a result of the coupling between the translational oscillation of the shell, in the acoustic potential well, and the shell centering force of the capillary wave, and needs to be investigated further.

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Table I

Properties of the Liquids Used in the Experiments at 25°C
** measured from the leftover flight fluids*
 # measured from retained samples of the flight fluids

Liquids	**υ (cSt)#**	**ρ (gm/cc)#**	**σ (dyn/cm)***
silicone oil (DC 200 series)	2.0	0.962	21±0.2
water	0.9	0.997	71.2±0.4
water/glycerine (72/28) (with 0.5% by volume green dye)	1.9	1.064	68.0±0.4

Interfacial tension between silicone oil (2 cst) and water/glycerine (72/28) is 33±0.3 dyn/cm#

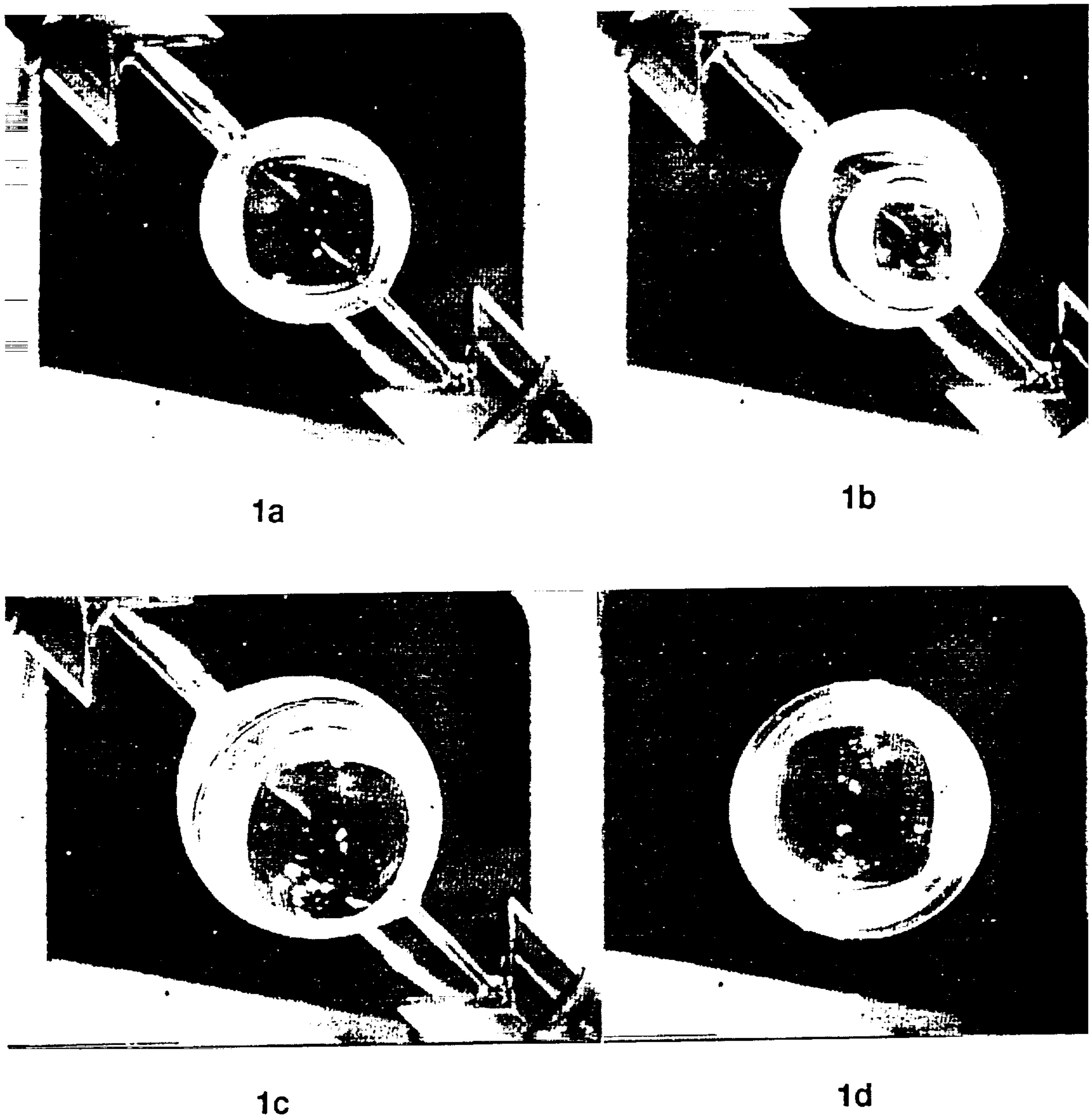


Figure 1 Deployment of a water shell (top-view): a) injected water drop held between needle tips, b) injection of air bubble, c) bubble injection complete, d) deployed water shell.

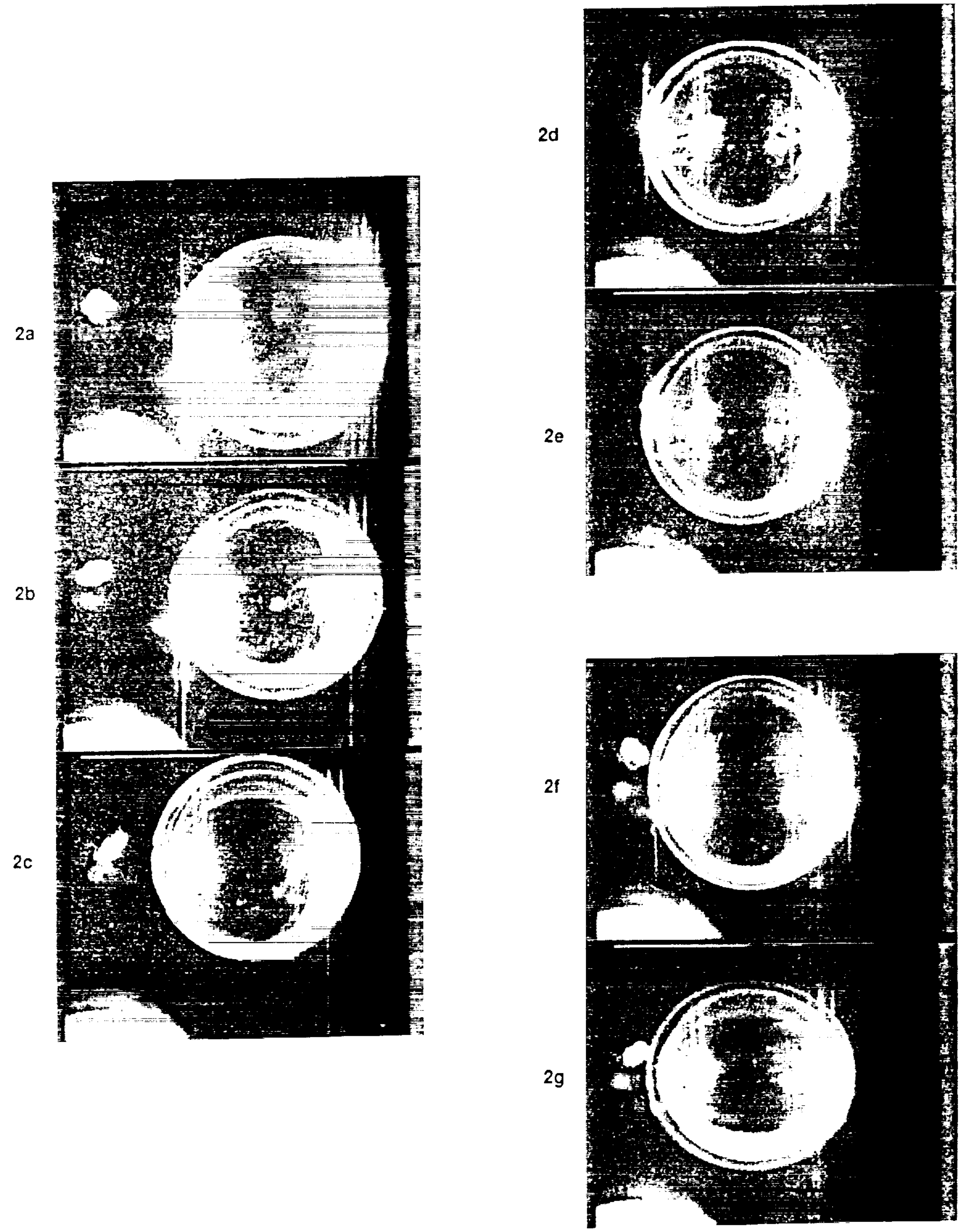


Figure 2 Centering of water shell by n=2 bubble mode: a, b, c) initial sloshing of the core due to translational oscillations of the drop in the potential well., d, e) core centered during small amplitude oscillations, f,g) centering during large amplitude oscillations.

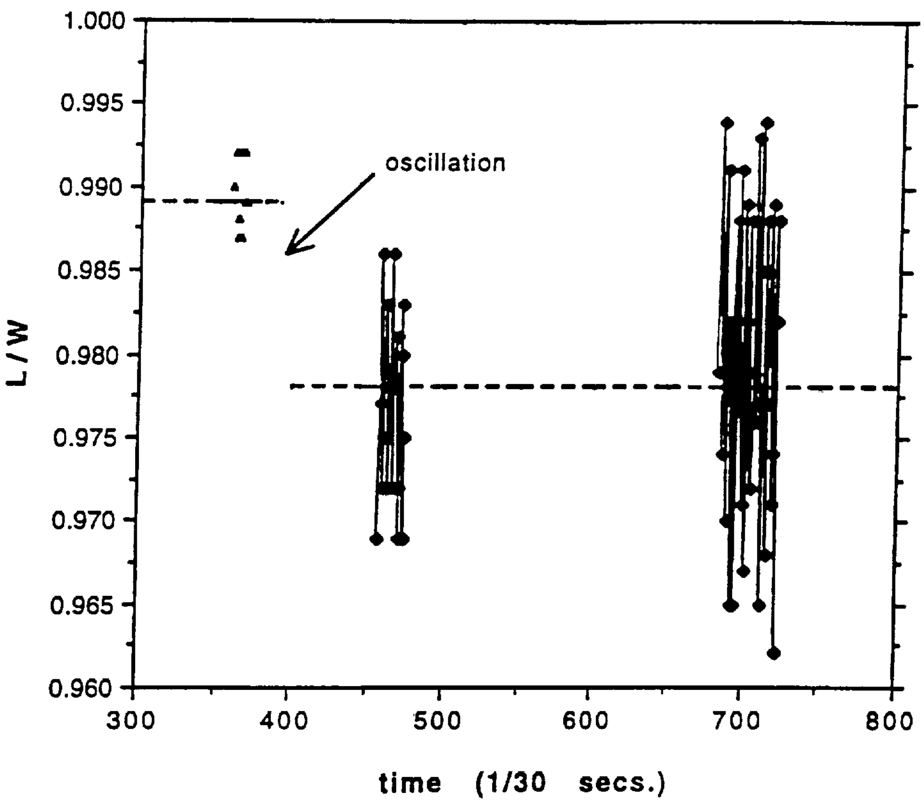


Figure 3 L/W versus time for n=2 bubble mode of the water shell.

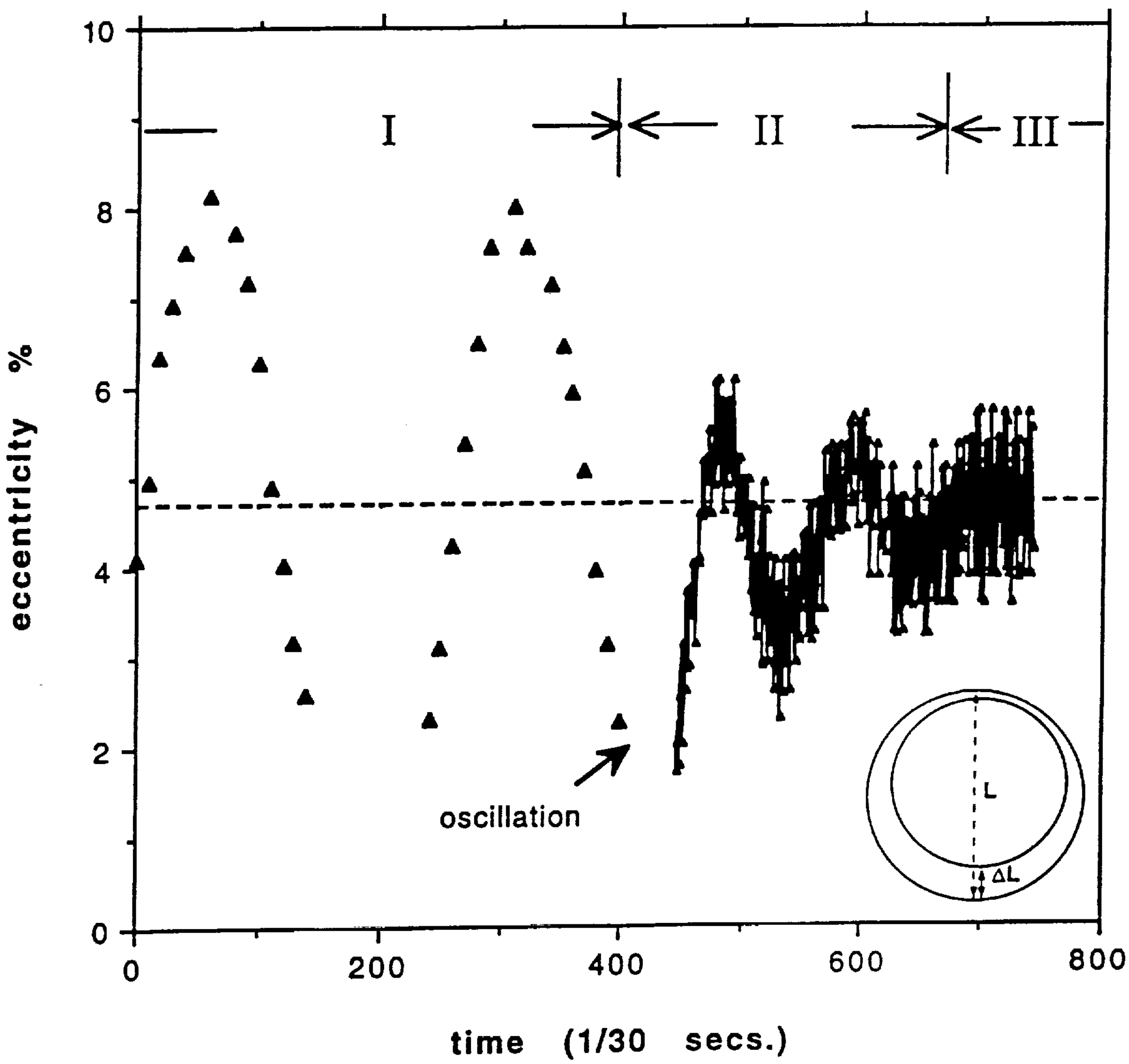


Figure 4 Plot of percentage eccentricity (ΔL/L *100) versus time for n = 2 bubble mode of the water shell.

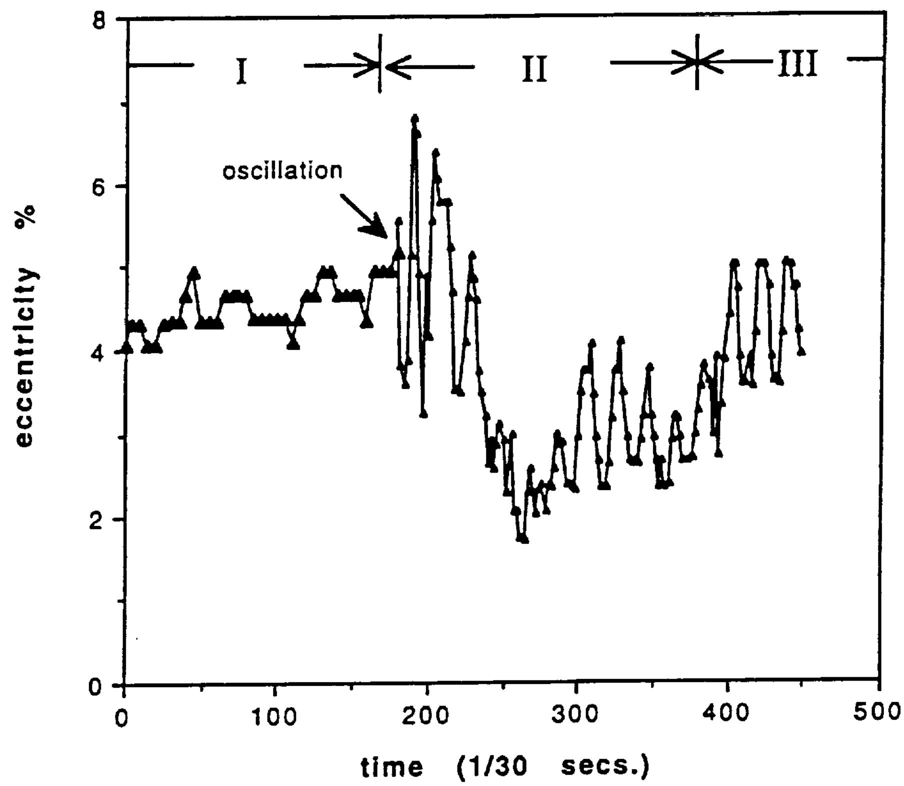
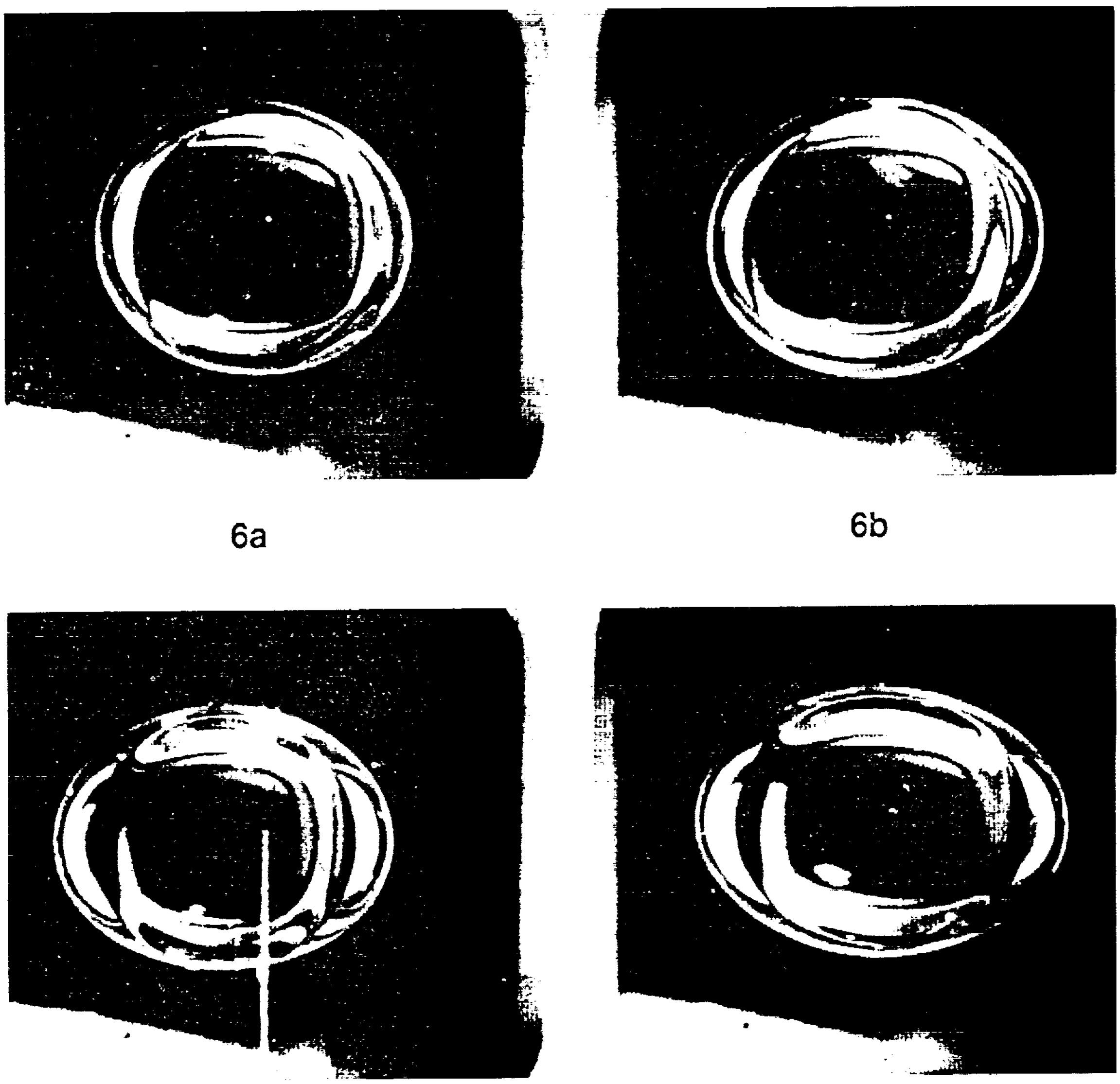
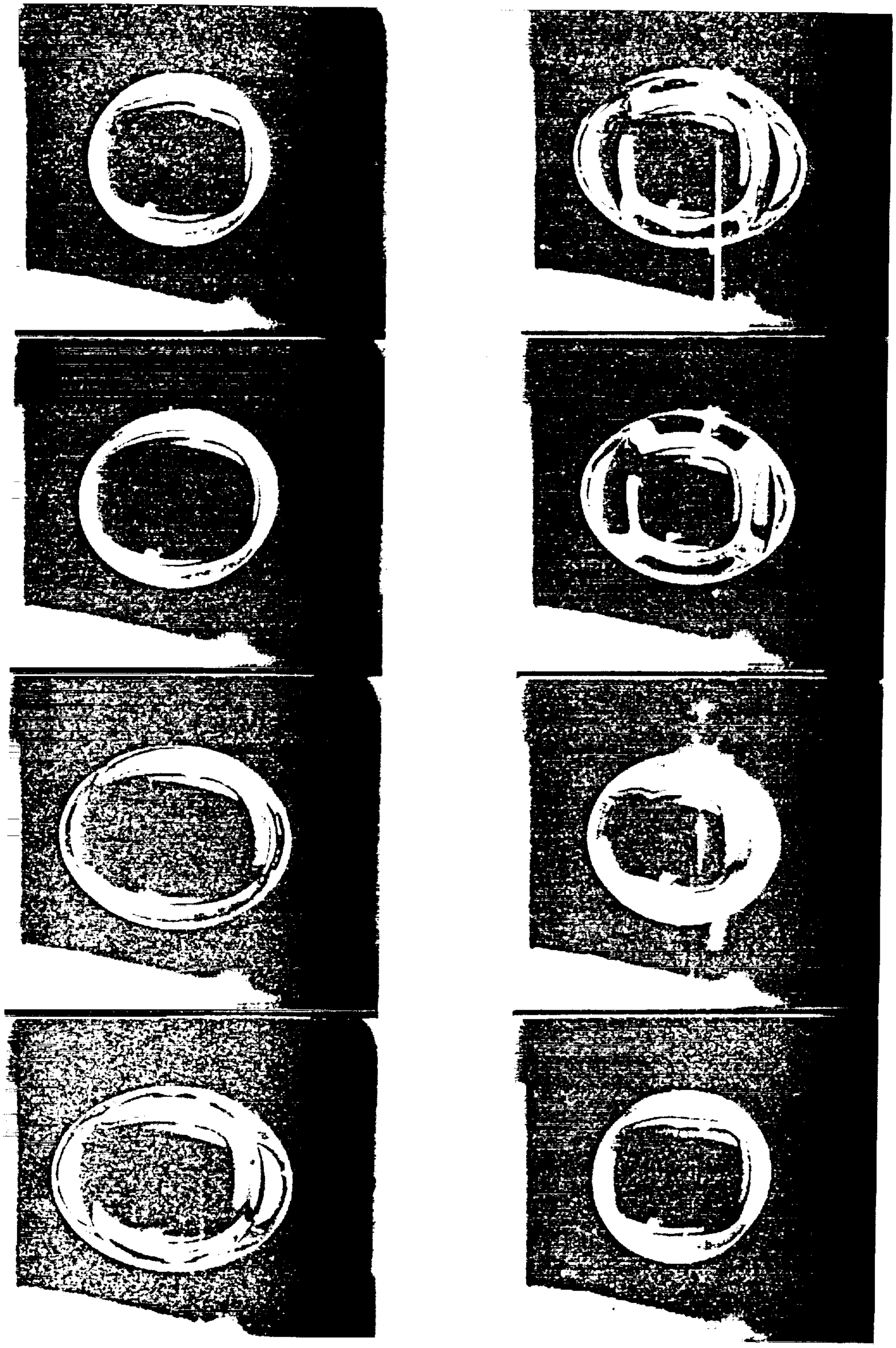


Figure 5 Plot of percentage eccentricity (ΔL/L *100) versus time for n = 2 slosh mode of the water shell.



6a 6b 6c 6d

Figure 6 Progressive centering of the liquid-core compound drop during n = 2 bubble mode oscillations with a strong m = ± 2 component (Figures a, b, c, d represent the shapes during the extremum of the oscillations).



7a 7e

7b 7f

7c 7g

7d 7h

Figure 7 A complete n = 2 bubble-mode oscillation following centering of the liquid core.

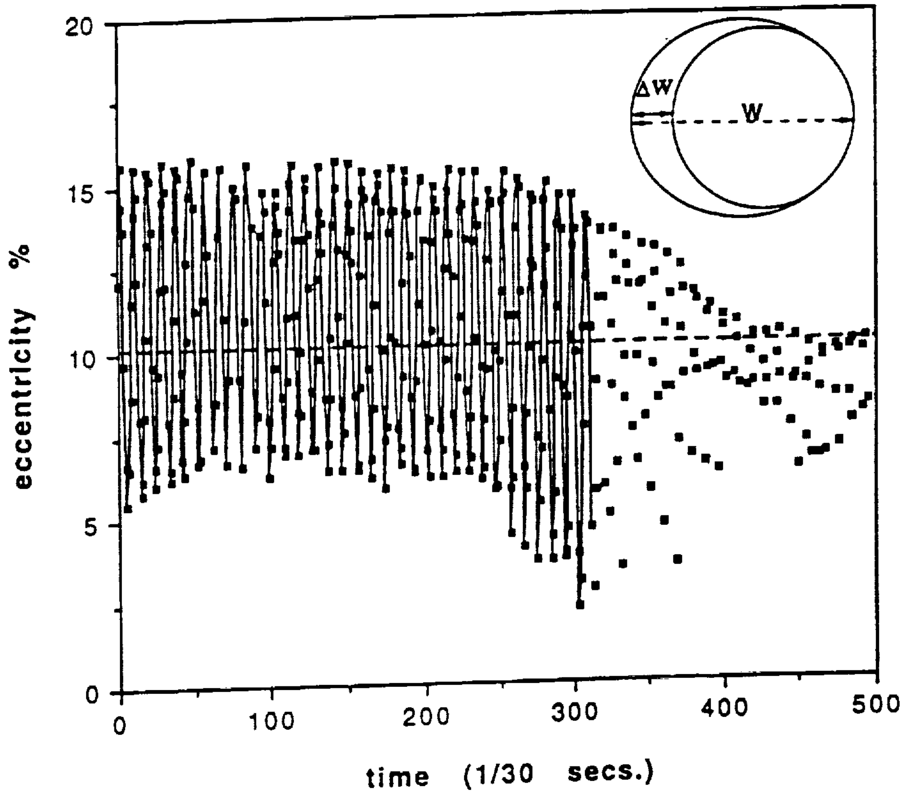


Figure 8 Plot of percentage eccentricity (ΔW/W *100) versus time for Figure 6 (a, b, c, d).

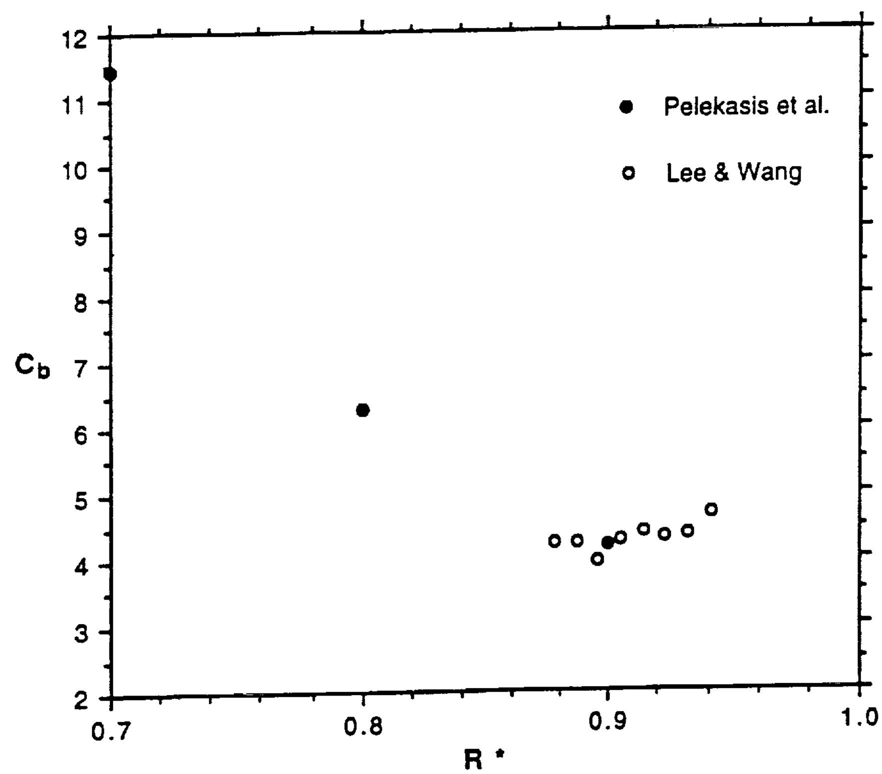


Figure 9 C_b versus R*. White squares: Lee and Wang 1988, black squares: Pelekasis et al. 1990.

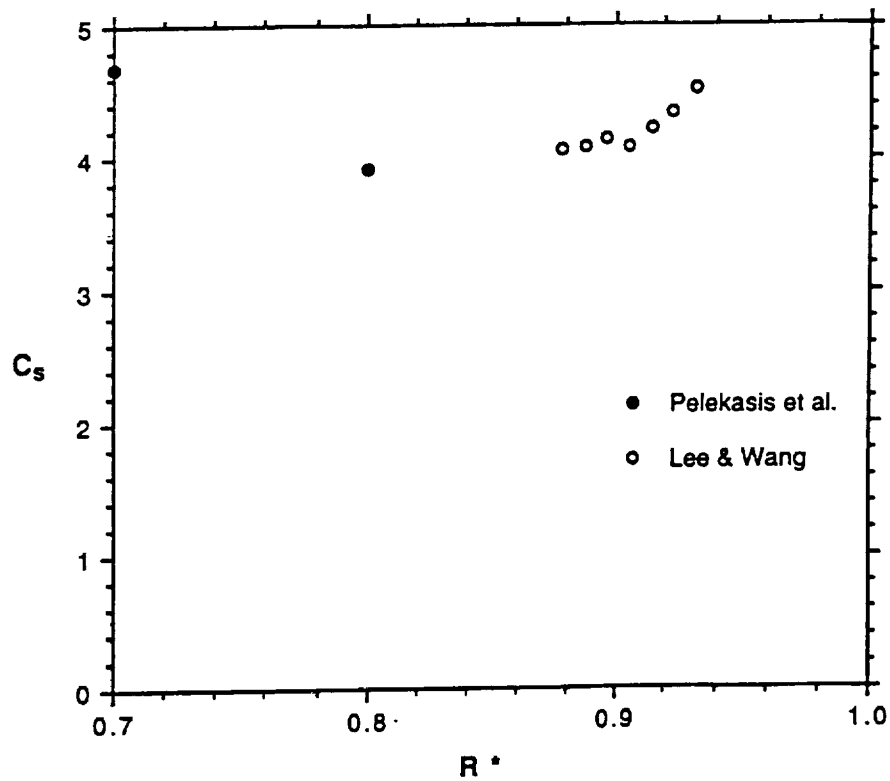


Figure 10 C_S versus R*. White squares: Lee and Wang 1988, black squares: Pelekasis et al. 1990.

Discussion

Question: *Could you comment on the interplay between viscous and inertial forces in the centering process?*

Answer: We don't know how to comment. We are looking at it. We think that the viscous force is very important. I believe that all the calculations currently assuming viscosity as a perturbation are probably incorrect. That is what the data suggests to us but not the calculations, but we are looking at it.

Question: *Wouldn't that be particularly important if the density difference is only a couple percent and could that sabotage your whole program ?*

Answer: It won't sabotage my program. We will just have to find a more ingenious way to do it, that's all. No. Actually the viscous force, we believe, manifests itself both in the liquid core as well as in the liquid shell. The data shows us that those are both equally important and if I had to draw a tentative conclusion, the tentative conclusion is the viscosity effect is very much stronger than predicted.

Question: *In the data you showed, where you did a compound liquid droplet what were the two liquids that you used and what were their viscosities ?*

Answer: Silicon oil, water, and glycerin.

Question: *In order to be able to see your data better why don't you color the inside droplet, so that when you are trying to do your data reduction you can see it a whole lot better ?*

Answer: We did, but it did not do any good on USML-1. We will try it again with a different color. We were using a green color and the sensor recordings did not come out very well. Part of the reason is that we have a lighting problem, the light was just not bright enough and that made life very difficult. During the investigation, I got a little bit involved in the medical-biotechnology and then I started to appreciate it. The human body is a remarkable system, particularly the eye and the eye could just barely make it out. We are hoping on USML-2 it will be better.