DROPS IN SPACE: SUPER OSCILLATIONS AND SURFACTANT STUDIES

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An unprecedented microgravity observation of maximal shape oscillations of a surfactant-bearing

water drop the size of a ping pong ball was observed during a mission of Space Shuttle Columbia as part

of the second United States Microgravity Laboratory-USML-2 (STS-73, October 20-November 5, 1995).

The observation was precipitated by the action of an intense sound field which produced a deforming force

on the drop. When this deforming force was suddenly reduced, the drop executed nearly free and

axisymmetric oscillations for several cycles, demonstrating a remarkable amplitude of nonlinear motion.

Whether arising from the discussion of modes of oscillation of the atomic nucleus, or the explosion of

stars, or how rain forms, the complex processes influencing the motion, fission, and coalescence of drops

have fascinated scientists for centuries. Therefore, the axisymmetric oscillations of a maximally deformed

liquid drop are noteworthy, not only for their scientific value but also for their aesthetic character.

Scientists from Yale University, the Jet Propulsion Laboratory (JPL) and Vanderbilt University

conducted liquid drop experiments in microgravity using the acoustic positioning/manipulation

environment of the Drop Physics Module (DPM). The Yale/JPL group's objectives were to study the

rheological properties of liquid drop surfaces on which are adsorbed surfactant molecules, and to infer

surface properties such as surface tension, Gibb's elasticity, and surface dilatational viscosity by using a

theory which relies on spherical symmetry to solve the momentum and mass transport equations [1]. The

technique involves the acoustic squeezing and releasing of the liquid drop, and the measurement of the

subsequent free decay frequency and damping constant [2]. For small amplitude motion, it is desirable to

excite only the lowest-order (energy) normal mode of the drop, the axisymmetric quadrupole mode, in

which the drop oscillates between an oblate and prolate shape. In our chamber this oblate-prolate alternation is seen in the X-view of the DPM, while in the Z view one observes a simple circular shape.

The Drop Physics Module (DPM) facility was designed by the Jet Propulsion Laboratory and built by Loral Corporation's Electro-Optical Sensors Division. This module is essentially an air-filled box at one atmosphere pressure with inner dimensions (X, Y, and Z) of 12.4 cm, 12.4 cm, and 15.2 cm, respectively. Four custom high-amplitude, titanium-dome acoustic loudspeakers were placed in the box along the intersections of the bottom and side panels. A pair of stepper-motor-controlled injectors are used to inject and retrieve drops, ranging from 1-14 cc in our experiments. A common configuration for drop manipulation consists of opposing driving speakers 1 and 3 in the (100) X mode, speakers 2 and 4 in the degenerate (010) Y mode, and all four speakers in the (001) Z mode, typically 1350, 1350, and 1130 Hz, respectively.

The payload crew scientists on board Columbia operated the DPM via an interactive software interface, allowing the alteration of the acoustical environment by changing speaker drive voltages, frequencies and phases. They were supported on the ground by the payload operations team of the Marshall Space Flight Center (Huntsville, Al.) and the DPM Science Team, which included the scientific investigators, development engineers and support personnel. Almost continuous real-time communication between the Spacelab team and the ground team via voice, telemetry, and video was available. These links enabled results to be immediately evaluated, allowing for parameter adjustments over the course of the experiments which could not be defined before the mission, and which were absolutely essential for the carefully timed sequences that led to our observations.

From that oscillation data both the frequency of oscillation and the decay constant can be retrieved. The oscillation is considered a "good one" if the Z view of the oscillation displays a circular outline for the drop which changes in size during the oscillation. This confirms the axisymmetric character of the oscillation. This aspect of the motion, along with the idealized spherical shape made possible by microgravity, makes data analysis straightforward.

The drop oscillation sequence reviewed here involved the oscillation of a 6.6 cc drop (2.33 cm diameter) water drop containing the non-ionic surfactant Triton-X-100, which is commonly used in detergents and mixing agents. The chemical formula is $CH_3C(CH_3)_2CH_2C(CH_3)_2C_6H_4E_{(n)}OH$, where $E=OCH_2CH_2$, and ranges from 9 to 10. It was saturated in the water at the critical micelle concentration

(CMC) — that is, the concentration beyond which small aggregates of the material will form in the bulk of the liquid and negligible lowering of the surface tension will occur. The CMC for Triton is a minuscule 1.4×10^{-4} g/ml, at which the approximate static surface tension of this aqueous solution is 0.03 Newtons/m (less than half that of pure water). It is thus readily apparent why these types of materials are of interest to scientists and engineers -- tiny concentrations leave bulk properties unchanged, but result in marked surface viscoelastic properties, and thus alter the surface and bulk motion dramatically.

The microgravity sequence (black background) shown in Figure 1 involved a drop that was slowly squeezed so that in the Z-view the circular outline grew and the X-view appeared initially as a narrow ellipse of aspect ratio of 4.5. One complete cycle is shown here, both in X and Z views, although the complete video shows about 20 cycles (in about 17 seconds) before the oscillation reverts to the lowest energy quadrupole mode. A World Wide Web animated video sequence can be found at <http://www.yale.edu/engineering/fac-info/apfel-data/drop.mpg>. The cigar-shaped image of 6th frame has an aspect ratio approaching 3, which, in the parlance of nuclear modeling is called "hyperdeformed" [3]. A second remarkable feature is that the drop did not fission. The lower surface tension that allowed for the large deformations also probably prevented the elongated sections from pinching off.

These observations are borne out by numerical computations of the evolution of the shape of greatly deformed drops using the boundary integral method as adapted by Tao et al. to the case of drop motion in the presence of an intense acoustic field which can be used to cause both static and dynamic shape changes.[4,5] This method permits the study of large free or forced shape oscillations of axisymmetric drops, including the effects of viscosity, but not including the presence of surfactants or surface damping. We wondered whether such an analysis would produce drop shapes comparable to our observations. If so, they would permit us to extract data such as the dynamic surface tension, which can differ from the statically measured value. Such differences can provide important information on the rate of processes occurring as surfactant transfers back and forth between the surface and its sublayer.

Figure 1 also shows the predictions of the shape oscillations using the boundary integral method (X-view). These frames are displayed above the corresponding observations. The numbers indicated in each frame show the nondimensional time given by the ratio of the real time to the factor $T = a_0/v_0$, where $v_0 = (2\sigma/\rho a_0)^{1/2}$, which is the capillary wave speed. Here σ is the surface tension, ρ is the liquid density, and a_0 is the drop radius. The computation was carried out for a Reynolds number ($\rho v_0 a_0/\mu$) of 600,

where μ is the shear viscosity, taken to be about 1.5 times that of water to account for the presence of the surfactant which increases the surface layer damping. The capillary wave speed is taken to be about 7.5 cm/s, which is estimated from the surface tension of Triton X-100 at this concentration. Note that when we also carried out the same calculation for zero viscosity, the predictions indicated that the drop would bifurcate, whereas in the case shown, the image tracks closely the experimental observations.

Analysis of the real time measurements from USML-2 and the non-dimensional time from the predictions of the boundary integral method allows one to compute a dynamic surface tension of approximately 33.5±1.0 dyne/cm (0.0335 N/m), which is slightly higher than the statically measured value for a 1 CMC solution of Triton X-100 of 31 dyne/cm. A higher dynamic surface tension might be expected, because surfactant transport to the surface is a rate-limited process, and the equilibrium concentration of surfactant is not fully achieved during the period of oscillation. While surfactant molecules in a thin boundary layer near the surface can reach the surface as it rapidly expands, diffusion from the bulk will be too slow to fully replenish the boundary layer. Yet, in the present case the drop begins in the statically deformed shape (maximum surface area), and therefore the surfactant does not have to diffuse to the surface, but only redistribute itself on the surface, which evidently happens on a time scale that is short compared to the drop motion.

It is truly remarkable that the shapes predicted correspond closely to the data. Such a correspondence through a complete cycle not only validates for the first time the use of the theoretical methodology of the boundary integral method for this special and unprecedent maximum oscillation observation, but also enables the prediction of dynamic surface tension . Future work will concentrate on the analysis of the decaying oscillation for this and one other surfactant material, which should permit the deduction of the surface viscosity coefficient and Gibbs elasticity, in addition to the dynamic surface tension.

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FIGURE CAPTION

Figure 1: Sequence of 14 frames showing single complete oscillation of a 6.6 cc drop that begins in a highly deformed (oblate) shape owing to a high intensity acoustic field, and which then oscillates after the acoustic field intensity is suddenly reduced. The top images in each row are the X (side) view, and the bottom images are the Z (top) view demonstrating the axisymmetric nature of the oscillation. The time of each frame is marked. Shown above the X view observations are a sequence of frames computed using the boundary integral method, assuming a Reynolds number of 600. The non-dimensional time, T, shown with each frame is defined in the text. Note that the last two frames of the predictions differ from the observations, because the prediction are showing the center plane of the drop (which is dimpled in), whereas the observations are showing the front view image which cannot show the inward dimpling effect.

t=0.00	t=0.55	t=0.75	t=0.95	t=1.25	t=1.80	t=3.05

X

X

Z

| t(sec) 0.00 | 0.083 | 0.113 | 0.143 | 0.193 | 0.283 | 0.463 |

t=3.50	t=3.75	t=4.10	t=4.55	t=4.95	t=5.10	t=5.45

X

X

Z

| t(sec) 0.533 | 0.563 | 0.613 | 0.683 | 0.743 | 0.783 | 0.843 |