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BIFURCATION OF ROTATING LIQUID DROPS: RESULTS FROM USML-1 EXPERIMENTS IN SPACE

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#### SUMMARY

Experiments on rotational bifurcation of liquid drops, in which the drops were levitated and spun using acoustic fields in a low-gravity environment, were conducted during the USML-1 Space Shuttle flight. The experiments have successfully resolved the discrepancies existing between the previous experimental results and the theoretical predictions. In the case of a spherical drop, for which theory exists, the results agree well with the predictions. In the case of flattened drops, the experiments have established a family of curves, with the spherical drop as the limiting case.

## INTRODUCTION

The phenomenon of bifurcation of a rotating liquid drop was first observed by Plateau (1863), as a model for a rotating liquid mass held by self-gravity. In his experiment, liquid drops were held and rotated by a shaft in a liquid medium of almost the same density. Beside the solid supporting device, his drops were necessarily influenced by the viscous friction with the outer liquid. We have performed the experiments for liquid drops in air without these limitations, using acoustic force for positioning and acoustic torque for rotation, in the microgravity environment of space.

When a spherical liquid drop, held together by surface tension, undergoes a solid body rotation about an axis in a less dense medium, its shape becomes oblate, with its liquid displaced away from the axis due to centrifugal force in the new equilibrium configuration. With increasing rotation rate, the equilibrium shape becomes more and more flattened, then becomes concave at the top and bottom surfaces, until eventually the two surfaces merge and the central membrane breaks. In reality, however, the drop finds a non-axisymmetric route to equilibrium, which leads to two-lobed (n=2) bifurcation, as the rotation rate reaches a certain critical value before the drop becomes concave (Chandrasekhar 1965). The n=2 bifurcation branch, of the maximum dimension versus rotation rate curve, has been calculated, among others, by Brown and Scriven (1980). With increasing angular momentum, as the drop

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approaches the other end of the bifurcation branch, at one point, the two-lobed shape turns from stable to neutrally stable, and then unstable, with the drop eventually fissioning.

The experiments have been conducted using acoustic fields. 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 (King 1934). It is also known that two sound fields vibrating in perpendicular directions, at the same frequency but out of 2-phase to generate a circular motion in the medium, can exert a viscous torque on the sphere (Busse and Wang 1981). These are the main principles behind the present apparatus as well as the Drop Dynamics Module (DDM) used in the previous space experiments (Wang et al. 1986).

In the previous space-based experiments, while there was a general qualitative agreement with the theory, there was also some discrepancy between the data and the theoretical curve which needs to be resolved. Rhim et al. (1988) performed the experiments in a ground-based study using charged drops levitated in an electrostatic field and rotated using an acoustic torque, obtaining an n=2 bifurcation point close to the theoretical prediction. But the use of electric charge and field introduces some uncertainty into the validity of the comparison. Biswas et al. (1991) studied the bifurcation of ground-based acoustically levitated and rotated drops in comparison with the 1986 space experiments. Although the outcome of their experiments was necessarily biased by gravity, their data has a signature that seems to corroborate that of the earlier space flight results. We shall address this point later.

For the bifurcation study, the results are conveniently displayed in a plot of the dimensionless radius R\* (R\* = R<sub>max</sub>/R) with the dimensionless rotation rate  $\Omega^*$  ( $\Omega^*=\Omega/\omega_0$ ) Here, R<sub>max</sub> is the maximum radial position of the axisymmetric or nonaxisymmetric drop surface in the equatorial plane from its central axis in gyrostatic equilibrium, R is the spherical radius of the drop,  $\Omega$  is the drop rotation rate and  $\omega_0 = (8\sigma/\rho R^3)^{1/2}$  is the n=2 oscillation frequency of the drop; with  $\sigma$  being the surface tension and  $\rho$  being the density of the drop liquid.

# I. EXPERIMENTAL APPARATUS AND PROCEDURE

The experiments were performed in the Drop Physics Module (DPM; Figure 1) 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, an injector for deploying drops consists of two similar arms sticking from the middle of two opposite vertical edges to meet at the center of the chamber when demanded, one of which contains the injector tube for the liquid while the other provides the mechanical means to hold the drop during the injection. Two types of liquids are used for the experiments: (1) silicone oil and (2) water-glycerine mixtures. Since they have different wetting properties, they are handled slightly

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differently. For deploying a silicone oil drop, a flat-tip injector is used such that a liquid column is sandwiched between two flat plates during injection (Figure 2). When the desired amount of liquid has been put into place, the arms are suddenly withdrawn, leaving the drop levitated at the center. The same procedure is used for water-glycerine drops, except that a needle-tip injector is used, such that the drop is held between two needle tips initially (Figure 3). A simple tip-to-tip deployment is the desired method, whereby a quiescent spherical drop can be deployed at the center of the chamber, and trapped by the acoustic potential well.

Since this is mainly a study of the solid body rotation of drops, transient dynamic situations such as those during the spin-up or spin-down process must be minimized as much as possible. Since the relaxation time for a drop to adjust to a new rotation rate is estimated to be  $R^2/v$ , where R is the radius of the drop and v is the kinematic viscosity of the liquid, high-viscosity liquids were used. The practical absence of gravity allows for the study of large drops, and the drop sizes are only limited by how much of a drop can remain in view through the windows while it is deformed by rotation.

Silicone oil is the best choice since, unlike water, for example, its surface tension is not very sensitive to contamination. This fact is important, considering that because of the flight planning, the liquids had to be prepared and stored for months before the flight. Pure water was also not used because of its low viscosity, which means long relaxation time. Glycerine/water (82/18 and 87/13 by weight) solution was used instead. The properties of the liquids used are listed in Table 1. Pliolite tracer particles (Goodyear Chemical Co.), about 50-100  $\mu$ m in size, are mixed with the liquids during preparation, for the purpose of identifying solid body rotation and determining the rotation rate. The volume of the drop is calibrated against a perfectly machined 2.54 cm plastic ball when the latter is held at the center of the chamber by the injector arms. The uncertainty in the volume determination is less than 3%.

For the bifurcation study, a drop is levitated and rotated about the z-axis using about 145 dB of sound pressure level along each of the axes. Minimum acoustic pressures are used to avoid deforming the drop. The rotation rate is increased stepwise a bit at a time, allowing plenty of time for the drop to relax in order to ensure solid body rotation. Experimentally, the drop stays at a fixed position and rotates steadily in gyrostatic equilibrium after its rotation rate has increased past a certain point; thereafter, usable data can be taken. Most measurements of the drop size  $R_{max}$  were made using the top-view (z-view), where the measurements were insensitive to any small movements of the drop. Most experiments were conducted at temperatures between 23.5° to 26.5° C. The average temperature for each of the drop, has been indicated below.

#### **II. RESULTS**

### A. Data from glycerine/water drop (82/1 8)

This solution was colored with a green food-coloring dye (McCormick), 0.1% by volume. The measurements shown in Table 1 for this 82/18 solution were made on the ground from the leftover samples.

In Figure 4 we show the data by plotting R\* versus  $\Omega^*$  for controlled spin-up and controlled spin-down of a 2.5 cc drop, where 'controlled' means 'changing the torque in small steps one at a time,' with the rotation rate scaled with the measured value of  $\omega_0$ . The experiment actually involved many spin-ups, spin-downs and bifurcations of the same drop, and the data points shown in Figure 4 represent the runs in which we are satisfied that the drop was in stable levitation and gyrostatic equilibrium. The axisymmetric curve lies slightly above, and the bifurcation branch lies slightly to the left, of the curves predicted by Brown and Scriven (1980).

In Figure 5 we show the data for a similar experiment with another drop in this session, with the drop significantly flattened by the z-drive during spin-up. The drop has a volume of 1.55±0.05 cc, and was deployed by a tip-to-tip injection between fresh tips (Figure 3). The average temperature during the experiments was 24°C. The deformation before the rotation started, defined as (a/b-1) x 100%, where a and b are the equatorial and polar radii, respectively, was about 25±2%. For the spin-up, the axisymmetric curve lies well above, and the bifurcation branch lies well to the left, of the predicted ones. The value of  $\Omega^*$  at bifurcation from the spin-up curve is about 0.47, which is significantly lower than the theoretical value. The bifurcation branch shoots vertically upward such that R\* increases by 10-15% at constant  $\Omega^*$  before bifurcation; it is noteworthy that bifurcation does not occur at the turning point but a bit above it. Thus the bifurcation point is not well-defined for such a drastically flattened drop. The bifurcation branch continues to rise vertically to intersect the theoretical bifurcation curve until fission occurs. The spin-down was achieved by lowering the z-drive close to (but slightly above) the x- and y-levels, and releasing the torque as opposed to a controlled spin-down. Since the torque was released when the drop was high up on the bifurcation curve, the drop, with its high viscosity, has enough time to relax back to equilibrium not too far down the curve. The rest of the spin-down curve and the associated bifurcation point agree well with the theory.

# B. Data from silicone oil drop (DC 200 series, 100 cst)

The first silicone oil drop had a volume of  $2.90\pm0.05$  cc. The average temperature during the experiment was  $22.5^{\circ}$ C (for which the surface tension is corrected from its value at  $25^{\circ}$ C in Table 1 at -0.08 dyn/cm/°C). The initial deformation of the drop due to the z-drive was about  $5\pm1\%$ . The torque was carefully raised in the spin-up. Starting from an axisymmetric shape, the drop went through bifurcation to the two-lobed shape, and then all the way to fission. As seen in Figure 6, the agreement with the theory

is good; except in the unstable two-lobed region where fission occurs. This issue will be taken up in the future.

The experiment was repeated with a second silicone oil drop of volume  $2.80\pm0.05$  cc at an average operating temperature of  $25^{\circ}$ C (Figure 7). In one run, the drop was spun step-by-step beyond bifurcation and then allowed to spin down with all acoustics turned off, starting from a point on the bifurcation curve about the same as that for the last run. The data, with all acoustics off, cannot be plotted because the dimension of the drop changed as it moved out of the field of view of the cameras in the absence of a restraining force. But the bifurcation point is estimated to be  $\Omega^* = 0.563$ , in good agreement with the theory. Finally, the drop was trapped and spun up past bifurcation to fission, with an initial flattening of  $11\pm1\%$ . As expected, there is a deviation from the theory as a consequence of the flattening (Figure 7).

In Figure 8, we show the photographs of the top view of a typical drop being spun up from rest, going through bifurcation to fission. During the fission (Figures 6,7), it is noted that  $R^{*2}\Omega^*$  is approximately constant, as is required by conservation of angular momentum.

The experimental uncertainty in  $\Omega^*$  is 2%, and that in R\* is 2%. It is necessary to flatten the drops to impose a controlled rotation; however, if the imposed deformation is less than 5%, the uncertainty for  $\Omega^*$  and R\* are within the aforementioned error bars.

### **III. COMMENTS**

The experiments have been successful in resolving the discrepancies existing between the previous experimental results and the theoretical predictions. In the case of a spherical drop, for which theory exists, the results agree well with the predictions. In the case of flattened drops, the experiments have established a family of curves, with the spherical drop as the limiting case.

In the previous Spacelab experimental results (Wang et al. 1986; cf. Fig. 3), for the spin-up and fission of a 1 cc glycerine-water drop of 100 cSt viscosity, the bifurcation branch is well to the left of the ideal one, and crosses it later. The value of  $\Omega^*$  at bifurcation is about 0.47±0.04, which is below the theoretical value of 0.56. Considering the qualitative similarity between these earlier results and the present results, shown in figure 5 for the flattened drop, and also those from the ground-based study using a single-axis levitator (Biswas et al. 1991), it is felt that the discrepancy in the earlier Spacelab results is due to drop flattening along the rotation axis by the acoustic radiation pressure. This has been confirmed by a re-examination of the initial non-rotating shape of the drop used in the earlier space experiments, the flattening being estimated to be about the same as that in the current Figure 5.

In a general context, a rotating drop with its 'elasticity' characterized by the n=2 oscillation frequency is not easily deformed by the acoustic pressure when it is still almost spherical. On the other

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hand, at the bifurcation point where the stability is neutral, the oscillation frequency is zero and the drop is 'plastic' rather than 'elastic.' Therefore as the drop flattens with rotation when approaching the bifurcation point, it also becomes more susceptible to deformation by the acoustic pressure. In other words, the bias shows up at higher rotation rate. The bias tends to make the drop flatter even if the three waves are of the same amplitude, because after the drop becomes oblate with rotation, it scatters more z-wave than others with its larger projection on the equatorial plane, and thus sees the z-wave as the dominant wave.

With such a bias, it is obvious that the axisymmetric curve should shift upward. Since the angular momentum balance of an axisymmetric drop is not affected by flattening, it seems reasonable to assume that the bifurcation angular momentum is insensitive to the flattening. Also, for the same angular momentum, the axisymmetric drop has a higher R\*, thus a higher moment of inertia and a lower  $\Omega^*$ . This possibly explains the leftward shift of the bifurcation point. Furthermore, when the drop becomes elongated, the acoustic radiation pressure continues to flatten it such that R\* rises more rapidly with decreasing rotation rate than the non-flattened counterpart. This possibly explains why the bifurcation curve for the flattened drop rises to intersect the ideal one on the  $\Omega^*$ -R\* plane.

Another question is why the bifurcation point from the ground-based study (Biswas et al. 1991) of about  $\Omega^*=0.44$  for an initial deformation of 21 % due to the levitating sound field, is lower than 0.47 for a drop of an even greater deformation of 25% in Figure 5, if we believe that the bifurcation point should go down with deformation. A reasonable explanation is that although gravity is much weaker than surface tension in the ground-based study, it can easily tip the energy balance at the bifurcation point, where surface tension is counteracted by centrifugal force to the point that the system is neutrally stable. Gravity tends to spread the drop out horizontally (like a drop on a table), increasing R\* and reducing  $\Omega^*$  for the same angular momentum.

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### Table 1

# Properties of the Liquids Used in the Experiments at 25°C

- \* measured from leftover flight liquids
- # measured from retained samples of the flight liquids
- ^ nominal values suggested by the manufacturer, or from standard tables

Liquids	ა(cSt)	ρ <b>(gm/cc)</b>	σ (dyn/cm)
Silicone Oil (DC 200 series)	100^	0.962#	21±0.2*
Glycerine/Water (82/18) (with 0.1% vol. green dye)	47^	1.209#	64.8±0.4*
Glycerine/Water (87/13)	87^	1.222#	64.4±0.4#



Figure 1 Schematic of the Drop Physics Module.

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Figure 2 Deployment of a silicone oil drop: a) top view showing the flat injector tips, b) the injected liquid column, c) the deployed drop, d) side view of the deployed drop.



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Figure 3 Tip-tip deployment of a glycerol-water drop: a)top view showing the needle tips, b) the injected liquid drop, c) the deployed drop, d) side view of the deployed drop.

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Figure 4 Bifurcation of 82/18 glycerine/water drop of 2.5 cc.



Figure 5 Bifurcation of 82/18 glycerine/water drop of 1.5 cc, the left curve was obtained by deliberately flattening the drop with the z-drive by 25%.

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Figure 6 Bifurcation of the silicone oil drop of 2.9 cc to fission.



Figure 7 Bifurcation of the silicone oil drop of 2.8 cc (the data points leading to fission was obtained when the drop was flattened with the z-drive by 11%).

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Figure 8 Photographs of the top view of a typical drop being spun up from rest, going through bifurcation to fission: a, b, c = axisymmetric shapes.



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Figure 8 (continued) Photographs of the top view of a typical drop being spun up from rest, going through bifurcation to fission: d, e, f, g = two-lobed shapes.



Figure 8 (continued) Photographs of the top view of a typical drop being spun up from rest, going through bifurcation to fission: h, i, j, k = necking and fission.

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