## A SYMMETRY BREAKING EXPERIMENT ABOARD MIR AND THE STABILITY OF ROTATING LIQUID FILMS

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## Abstract

We discuss results from two parts of our study on the behavior of liquids under low-gravity conditions. The first concerns the Interface Configuration Experiment (ICE) aboard the Space Station Mir on the Mir-21/NASA-2 mission; for a certain "exotic" container, distinct asymmetric liquid configurations are found as locally stable ones, even though the container itself is rotationally symmetric, in confirmation of mathematical results and numerical computations. The second investigation concerns the behavior of slowly rotating liquids; it is found that a rotating film instability observed previously in a physical experiment in 1-g, scaled to render gravity effects small, does not correspond to mathematical and computational results obtained for low-gravity. These latter results are based on the classical equilibrium theory, enhanced with a van der Waals potential of adhesion.

### **1** Introduction

It is essential, when planning space-based operations, to be able to predict the configurations that fluids will assume in their containers under low-gravity conditions. In such an environment surface wettability and container geometry predominate in governing equilibrium behavior. As formulated in the classical Young-Laplace-Gauss (Y-L-G) theory of capillarity, the wetting characteristics of a particular system are embodied in a well-defined equilibrium macroscopic contact angle depending only on the materials (see, e.g., [10, Chap. 1]).

According to the Y-L-G formulation, the mechanical energy E of a partly filled container, when gravity is absent or can be neglected, the situation we discuss primarily, is

$$E = \sigma(S - S^* \cos \gamma). \tag{1}$$

Here  $\sigma$  is the liquid-gas interfacial tension and  $\cos \gamma$  is the liquid-solid-gas relative adhesion coefficient,  $\gamma$  being the contact angle between the liquid and the container; Sdenotes the area of the liquid-gas free surface and  $S^*$  the area of the portion of the container in contact with the liquid. Configurations of the liquid that yield a stationary value for E subject to the constraint of fixed liquid volume are, according to the classical theory, equilibrium configurations. They will be stable, as well, if E is locally minimized

Even under the simplifications of zero gravity or lowgravity conditions for which gravitational energy can be neglected, the strongly nonlinear governing equations from the classical theory are difficult to solve. Closedform solutions for this case have been obtained for only a few simple configurations, such as in right circular cylindrical or spherical containers. Departures from these container geometries can lead to dramatic changes in fluid behavior [6], [10].

We discuss here results from two parts of our study on behavior of liquids under low-gravity conditions. The first concerns a space experiment that investigates actual physical behavior for a situation in which symmetrybreaking type of mathematical results were obtained for equilibrium behavior based on the classical Y-L-G formulation. The second concerns computational results for uniformly rotating highly wetting liquids, based on the Y-L-G formulation enhanced with a van der Waals potential of adhesion.

We are interested especially in observing the extent to which some of the striking predictions of the fully nonlinear Y-L-G theory will be observed experimentally in the presence of factors not reflected in that theory, such as contact angle hysteresis and dynamic contact line phenomena. In coordination with this effort, we seek indications as to the effects these factors will have on fluid behavior. We are guided in our procedures to some extent by the previous experiments recorded in [9], in which a discontinuous behavior predicted by the Y-L-G theory was in fact observed. The mathematical, computational, and pre-flight drop tower experiment background for those experiments can be found in [4]; other examples of such experiments can be found in the references of [4] and also in [6].



Figure 1: Axial section of exotic container, with members (dashed curves) of continuum of rotationally symmetric equilibrium interfaces, all of which meet the container with the same contact angle  $\gamma$  and enclose the same volume V of liquid with the container.

### 2 Exotic containers

## 2.1 Background

Of interest here are the "exotic" containers discussed in [7] from a theoretical point of view (based on the Y-L-G theory). These containers are rotationally symmetric with the remarkable property that for given contact angle and liquid volume, an infinity (in fact, an entire continuum) of distinct rotationally symmetric equilibrium configurations can appear, all of which have the same mechanical energy. Such container shapes, first studied in [12] for the special case of zero gravity and  $\gamma = 90^{\circ}$ , are possible for any gravity level, but only for low-gravity conditions can the phenomena be expected to appear on an adequately large size scale to be readily measurable. For zero gravity the size scale can be arbitrary. Additionally, these containers exhibit marked symmetry-breaking properties, as discussed below.

The axial section of an exotic container for zero gravity and contact angle 80° is depicted in Fig. 1. The mathematically derived "exotic-bulge" portion is joined to circular cylindrical extensions and disc ends to form a closed container. The dashed curves depict members of the continuum of rotationally symmetric equilibrium interfaces, all of which enclose the same volume V of liquid with the bottom and walls of the container. The horizontal planar free surface  $\mathcal{P}$  making the specified contact angle with the container is of necessity a member of the family. The topmost curved interface, which is discussed below, is denoted by  $\mathcal{T}$ .

It turns out that the rotationally symmetric equilib-

rium configurations are unstable; as is shown in [11] and [5], particular deformations that are not rotationally symmetric yield configurations with lower energy. Thus, it is possible to demonstrate a symmetric container that admits infinitely many symmetric equilibrium interfaces, but for which no interface that minimizes energy can be symmetric. This is in notable contrast with what happens in the familiar case of the right circular cylinder, for which the symmetric interface (unique for prescribed liquid volume) is stable, and no asymmetric ones can appear.

There is presently no known way to determine mathematically the surfaces that minimize energy in the exotic containers. However, numerical computations have suggested a number of particular non-rotationally-symmetric surfaces as local minima. Shown in Fig. 2 are three distinct non-rotationally-symmetric local energy minimizing surfaces for an exotic container, as obtained numerically in [3] using a modification of an early version of the Surface Evolver software package [1]. These surfaces, all of whose energies are less than that of the symmetric equilibrium family's, are depicted in order of increasing energy from left to right. We refer to the surface configurations as the "spoon", the "potato chip", and the "lichen". The contact lines of these surfaces with the container undergo respectively one, two, and three excursions from upper to lower extremities and back as they circumnavigate the bulge.

## 2.2 Space experiment

Depicted in Fig. 3 is the vessel for the Interface Configuration Experiment (ICE) carried out during the Mir-21/NASA-2 mission on board the Mir Space Station. Space flight experiments of such vessels on the carlier USML-1 Spacelab mission confirmed that the spoon shape (Fig. 2) is a strongly stable configuration, apparently a global energy minimizer. However, the other two nonaxisymmetric locally energy minimizing interfaces of Fig. 2 did not appear in that mission. Specific goals on Mir include an attempt to observe one of those configurations, the potato chip, and to test its stability.

To begin the experiment, which was carried out in the Mir glovebox module, a crew member attaches the ICE vessel on its side (the configuration in Fig. 3) to a labjack that is held to the glovebox floor with magnets. The reservoir valve is then opened, and the control dial is turned to displace the entire contents of the reservoir into the exotic container portion of the vessel. After a sufficient time, disturbances are imparted to the vessel and the resulting interfaces are observed, generally until they

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Figure 2: Computed locally stable configurations in an exotic container for zero gravity and  $\gamma = 80^{\circ}$ . From left to right, in order of increasing energy, "spoon", "potato chip", and "lichen". All have smaller energy than the symmetric equilibrium family.



Figure 3: Experiment vessel.

become quiescent again. The disturbances are increased in amplitude until the interface either consistently returns to a particular configuration or breaks up. The two video cameras recording the process are mounted for orthogonal views, and audio is recorded simultaneously.

After the fluid was carefully dispensed into an exotic container in a manner that encouraged establishment of a configuration like that for the topmost member of the family of equipotential rotationally symmetric surfaces ( $\mathcal{T}$  in Fig. 1), the crew member would then impart a perturbing impulse to the liquid by tapping the outside of the vessel with her finger. The solitary taps began as light disturbances, allowing for complete stabilization of the interface between each tap. These lighter taps were intended to dislodge a surface that might otherwise be unstable, but was "sticking" in its configuration because of resistance at the contact line (hysteresis). Subsequently, intensity of the taps was increased to test the stability of any configuration that might have formed. To induce larger amplitude axial disturbances, the crew member could slide the vessel-labjack assembly back and forth along the glovebox floor. These larger disturbances were intended to induce the liquid to leave a locally stable configuration and to settle into some different one.

Two distinct locally stable nonaxisymmetric interfaces were observed in the experiment, a spoon and a potato chip. Four equilibrium interfaces in all were formed, as shown in Fig. 4. The first was a rotationally symmetric interface (cf., T in Fig. 1) when the fill procedure was close to complete. It soon destabilized after completion of the fill, as a result of small disturbances imparted by the crew member in removing her hands from the vessel. Then an apparent global energy minimizer formed, a spoon like that found computationally and on USML-1, which configuration was stable to additional perturbing disturbances. Following further carefully applied moderately large disturbances another local minimizer formed, a potato chip like that in Fig. 2. This too was stable to additional perturbing disturbances. Finally, a further sizable disturbance led once more to the spoon, this time in reflected configuration.

It is important to remark that in the experiment the mathematical requirements for an exotic container are not perfectly met. There is of necessity a deviation of parameters (contact angle, liquid volume, gravity level) from their exact exotic values. Nevertheless, the asymmetry of the local minimizers did occur. This is in accord with computational evidence that the asymmetric configuration phenomenon generally is robust with respect to discretization errors from numerical approximation. Additionally, it was observed in the experiment that even with moderately large departures from the exotic requirements (70° liquid in a 55° exotic container) the asymmetric local-minimizer property persists.

Even if conditions for an exotic container are closely satisfied, once the fluid becomes asymmetric the presence of symmetric equilibria nearby the initial one probably has little bearing on the fluid's subsequent behavior. In the experiment, the presence of the continuum of distinct symmetric equilibria apparently has its main effect

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Figure 4: Configurations from Mir experiment. From left to right: (pinned) rotationally symmetric interface, spoon-right, potato chip, spoon-left

just in rendering the initial symmetric configuration unstable. It is of interest to note that in the mathematical proof of instability of the exotic continuum family only the local boundary curvature enters; if the curvature is greater than a certain lower bound, then the interface can be shown to be mathematically unstable, whether or not the container is exotic. (See [5], [11]; a proof of instability that extends to all gravity levels will appear in [15].)

## 2.3 Discussion

What began for us as a study of nonuniqueness and of exotic containers has led to our encountering the striking phenomenon of the asymmetry of energy minimizing liquid configurations in containers that are rotationally symmetric and for which the prescribed boundary data (contact angles) are also symmetric.

The accumulated evidence of this experiment and of its predecessors suggests strongly that in many cases of interest the Y-L-G theory reliably predicts fluid behavior in low gravity environments, or otherwise when surface forces predominate. Although in some cases considerable time (perhaps on the order of hours) may be required to overcome the effects of contact angle hysteresis, the fluid configurations finally adopted do appear to conform to predictions of the theory. The evidence suggests additionally that only in particular, perhaps rare, circumstances will capillary surfaces be uniquely determined by the data. Nor can it be expected that symmetric data will lead to symmetric configurations. In fact, as discussed above, even though the specifications for the exotic containers cannot be met exactly in practice, locally stable asymmetric surfaces will appear in them and will appear even for configurations that are distant from being exotic. These results can be important to designers of fluid management systems in low-gravity. The experimental results lend support to use of the classical Y-L-G theory in such design procedures, and it must be noted that striking and unexpected behavior predicted by that theory will actually be observed in practice. Full details of the experimental results for ICE, including data on time-dependent dynamic behavior, are given in [8].

## 3 Highly wetting, slowly rotating liquids

The results discussed above apply to equilibrium configurations and to contact angles within the range of applicability of classical versions of the Y-L-G theory. In this section we discuss a special configuration for which straightforward formal use of the basic premises of that theory may be insufficient.

As an example, consider a circular cylindrical rod that is dipped into a reservoir of wetting liquid. According to the classical Y-L-G theory, the liquid will move part way along the rod to a distance that is bounded for all contact angles, even 0°. For a "super-wetting" liquid, the type we wish to consider here, liquid will move over an additional portion, typically all, of the rod's surface coating it with a film. If the entire configuration is rotated about the rod's axis, the difference between the classical and highly wetting cases could give rise to very different behavior. In the first case the fluid will simply shift toward the outer portion of the reservoir, retreating somewhat along the rod, reaching a new quasi-steady equilibrium configuration. In the highly wetting case, however, it may happen that the film on the rod becomes unstable. Which case occurs can be crucial in certain applications, such as for slowly rotating liquid helium cryogenic cooling devices for orbiting satellites.

A coaxial cylindrical configuration has been proposed for such a cooling device for use in conjunction with the STEP and Gravity Probe-B Relativity Mission studies. A central rod, which contains experiment apparatus, is coaxial with a surrounding circular cylindrical

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Figure 5: Axial section of rotating fluid configuration.

container. The region between the rod and the outer cylinder is partially full of liquid, and the entire configuration is to rotate with uniform speed  $\omega$  such that any gas bubble is wrapped symmetrically about the rod, thereby keeping the center of mass on the axis, as desired for the experiments (Fig. 5). Using the Surface Evolver software [1], Brakke calculated configurations that a single bubble would have, based on the classical Y-L-G formulation for zero gravity for a liquid with zero degree contact angle [2]. He found that if  $\omega$  is not too small, then a gas bubble will assume the desired axisymmetric shape wrapped around the rod, as depicted in Fig. 5. The liquid makes a zero degree contact angle with the rod and does not wet the rod's central portion.

In a subsequent 1-g experimental study, which attempted to minimize the relative effects of gravity (by using small physical dimensions and large rotation speeds), it was found that for the highly wetting liquid used (silicon oil) a configuration similar to that of Fig. 5 developed, except that a liquid film formed over the rod covering the otherwise exposed central portion. The film became unstable; liquid was "pumped" continually from the bulk of the liquid into the film, with blobs of liquid departing from the film sporadically. A steady configuration never was achieved [14].

In a subsequent mathematical study with the purpose of providing a theoretical framework for the experimental results, a Y-L-G formulation was used, modified to allow for the presence of a film on the rod [13]. In this modified formulation one replaces the term  $-\sigma S^* \cos \gamma$ in (1), corresponding to the contact energy of the wetted and dry portions of the rod, with a van der Waals potential of adhesion  $-\alpha/(r - r_0)^3$  integrated over the liquid volume, with  $\alpha > 0$ ,  $\alpha$  small, an adhesion coefficient. The rod, of radius  $r_0$ , is covered by the film, and r is the radial coordinate of a point on the gas-liquid interface. Additionally, a rotational potential energy  $-(\rho \omega^2/2)r^2$ integrated over the liquid volume is added to the right of (1).

For a model problem of a rotating cylindrical rod covered with a film of uniform thickness, the analysis led to the interesting result that there is a critical value, depending on the radius of the rod, the liquid parameters, and the rotation rate, such that any film of thickness less than the critical value is stable, no matter how long the rod. On the other hand, if the film is thicker than the critical value, then there is a critical length l depending on the above parameters and the film thickness, such that all cylindrical films shorter than l are stable while all longer films are unstable (see Fig. 6). By comparison, for the classical Plateau-Rayleigh instability for a cylindrical column composed entirely of liquid, there is a critical length for any prescribed volume of liquid; a column is unstable for lengths longer than the critical, while it is stable for shorter lengths.

Numerical experiments based on the enhanced formulation were carried out for the actual container geometry using the Surface Evolver. It was found, over a broad range of parameters that include those of practical physical interest, that the liquid films that form on the central rod were always sufficiently thin so as to remain stable, never becoming thick enough to satisfy the mathematical criteria for instability. In fact, one can show analytically that an energy minimum occurs for film thickness  $O(\alpha^{1/3})$  asymptotically for small  $\alpha$ ; this result can be used with other asymptotic estimates to underpin the numerically found film stability. The numerical results indicate additionally that the ratio of the critical film thickness to  $\alpha^{1/3}$  is nearly constant for a large range of  $\alpha$ , lending additional support to the stability conclusion. Complete details are to be presented in a paper currently in preparation.

We conclude from the numerical study that the experimentally observed liquid film instability must derive from factors other than the above Plateau-Rayleigh type one. It is not known presently if an experiment conducted in low-gravity, free of the scaling and complications inherent in a 1-g experiment simulation, would exhibit the instability.

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Figure 6: Stable and unstable films for film thickness greater than critical value.

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