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# NONLINEAR DYNAMICS OF DROPS AND BUBBLES AND CHAOTIC PHENOMENA

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## ABSTRACT

Nonlinear phenomena associated with the dynamics of free drops and bubbles are investigated analytically, numerically and experimentally. Although newly developed levitation and measurement techniques have been implemented, the full experimental validation of theoretical predictions has been hindered by interfering artifacts associated with levitation in the Earth gravitational field. The low gravity environment of orbital space flight has been shown to provide a more quiescent environment which can be utilized to better match the idealized theoretical conditions. The research effort described in this paper is a closely coupled collaboration between predictive and guiding theoretical activities and a unique experimental program involving the ultrasonic and electrostatic levitation of single droplets and bubbles. The goal is to develop and to validate methods based on nonlinear dynamics for the understanding of the large amplitude oscillatory response of single drops and bubbles to both isotropic and asymmetric pressure stimuli. The first specific area of interest has been the resonant coupling between volume and shape oscillatory modes of isolated gas or vapor bubbles in a liquid host. The result of a multiple time-scale asymptotic treatment, combined with domain perturbation and bifurcation methods, has been the prediction of resonant and nearresonant coupling between volume and shape modes leading to stable as well as chaotic oscillations. Experimental investigations of the large amplitude shape oscillation modes of centimeter-size single bubbles trapped in water at 1 G and under reduced hydrostatic pressure, have suggested the possibility of a low gravity experiment to study the direct coupling between these low frequency shape modes and the volume pulsation, sound-radiating mode. The second subject of interest has involved numerical modeling, using the boundary integral method, of the large amplitude shape oscillations of charged and uncharged drops in the presence of a static or time-varying electric field. Theoretically predicted non linearity in the resonant frequency of the fundamental quadrupole mode has been verified by the accompanying experimental studies. Additional phenomena such as hysteresis in the frequency response of ultrasonically levitated droplets in the presence of a time varying electric field, and mode coupling in the oscillations of ultrasonically modulated droplets, have also been uncovered. One of the results of this ground-based research has been the identification and characterization of phenomena strictly associated with the influence of the gravitational field. This has also allowed us to identify the specific requirements for potential microgravity investigations yielding new information not obtainable on Earth.

#### I. INTRODUCTION

The application of the methods of nonlinear dynamics to the problem of the oscillatory behavior of single drops and bubbles has provided a new, more integrated framework for the analysis of the very diverse phenomena that are observed over a wide range of experimental parameters. It also underscores the similarities as well as the fundamental differences found in the large amplitude dynamics of drops and bubbles. A resurgence of interest in this area has taken place in recent years due to a combination of circumstances involving the development of improved numerical techniques, the introduction of novel experimental methods based on single particle levitation, and the recognition of a number of new application areas. The opportunities offered for experimentation in low gravity and the accompanying institutional support have also greatly contributed to the recent advances in this discipline. The ability to observe and to quantitatively measure the dynamical variables associated with the motion of isolated drops and bubbles has added, and will continue to add, to the data base that is necessary to induce new theoretical effort, as well as to compare with existing predictions.

Models for acoustically driven spherical gas bubbles in water have been analyzed at resonance conditions with the methods of chaos physics 1,2 to reveal a recurrent pattern in the bifurcation sets characteristic of period doubling cascades to chaos. Direct experimental evidence of chaotic radial oscillations has, however, not yet been obtained<sup>3</sup>. On the other hand, periodic, quasiperiodic, and chaotic light emission from sonoluminescing bubbles has been experimentally discovered <sup>4</sup>. Interest in the coupling between the low frequency shape oscillatory modes with the radial sound emitting oscillations of bubbles in water has arisen due to the possibility of previously unrecognized contributions to ambient sea noise, for example from gas bubbles entrained in breaking waves, as suggested by Longuet-Higgins <sup>5,6</sup>. Although experimental evidence for sound radiation from bubbles generated from the impact of droplets on a free liquid surface exists <sup>7,8</sup>, no verification of the direct excitation of radial bubble oscillation from shape modes has yet been provided. Prior theoretical treatments of the radial to shape mode coupling 9,10 have yielded evidence for chaotic bubble oscillations through a period doubling sequence (reference 9), and for an inviscid fluid through a homoclinic orbit (reference 10). The recently developed capability of ultrasonically trapping centimeter-size bubbles in water and of inducing large amplitude higher mode number shape oscillations 11,12, has been used to investigate the shape to radial mode coupling mechanism. The first part of this paper describes the theoretical and experimental studies motivated by the definition of a potential low gravity experiment that is aimed at the direct observation of this coupling.

Large amplitude shape oscillations of single drops have been extensively investigated in recent years through theoretical as well as experimental means. Early experimental results in the linear and nonlinear regions of oscillating drops <sup>13</sup> were motivated by the results of numerical studies using marker and cell techniques <sup>14</sup> as well as by the upcoming performance of a low gravity flight experiment <sup>15</sup>. Subsequent numerical treatments of nonlinear drop shape oscillations have considered the stability and mode coupling of inviscid electrically uncharged <sup>16</sup>, as well as charged drops <sup>17</sup>. More recent finite element treatments of the dynamics of freely suspended drops have been provided by Lundgren and Mansour <sup>18</sup> and Basaran <sup>19</sup> among others. The presence of a steady and time varying electric field has also been studied by Feng and Beard <sup>20</sup> and Kang <sup>21</sup>. Inviscid and low viscosity liquid behavior have thus been numerically simulated, and some limited experimental evidence for nonlinear behavior of droplets acoustically levitated in a liquid host was provided by Trinh and Wang<sup>22</sup>, and for drops in a gas by Becker et al.<sup>23</sup> and Trinh et al.<sup>24</sup>. Additional experimental data have been gathered in low gravity using acoustically positioned drops during a fourteenday Space Shuttle mission in 1992. In the second part of this paper, we describe a numerical study using boundary integral methods which deals with the shape and oscillations of a free drop in the presence of a constant as well as time-varying electric field. Recent experimental results obtained with the use of a hybrid ultrasonic-electrostatic drop levitator will also be discussed.

Before dealing with the nonlinear dynamics of free bubbles and drops, we will present some of the recent findings obtained from microgravity experiments. The purpose of this short summary is the identification of the principal differences between Earth-based and orbit-based experimental conditions.

# **II. SOME RESULTS OF LOW GRAVITY BUBBLE AND DROP EXPERIMENTS**

#### A.Drop experiments in microgravity.

A set of experiments on USML-1 dealt with the equilibrium shape of rotating free drops. Similar acoustic levitation and rotation methods were used in Earth-based and Spacelab experiments. Because of the static distortion, unavoidable with uncharged levitated drops in 1 G, theoretical predictions could not be verified. The onset of bifurcation was always measured at lower rotation rate than predicted. A parametric study with the value of the static distortion as the variable showed that the measured bifurcation point decreases with increasing oblate static deformation  $^{25}$ . As shown in figure 1, the removal of this static shape distortion in the space experiment allowed the verification of the theoretical prediction.

B. Bubble oscillations in low gravity.

A small ultrasonic device for bubble dynamics studies was flown aboard the Space Shuttle during the USML-1 Spacelab mission in 1992. One of the main results was the verification of the drastic reduction in

static shape distortion and the elimination of capillary wave excitation at the bubble surface by the ultrasonic wave. As figure 2 indicates, the results of the measurement of the fundamental mode frequency for large bubbles in 1 G deviates from the theoretically expected values. Measurement in low gravity removes this discrepancy for both the fundamental and the next higher mode. Thus, the most obvious manifestation of the interfering effects of the positioning and manipulating ultrasonic field found in 1 G is eliminated because the intensity of this field is greatly reduced in microgravity.

# **III. NONLINEAR BUBBLE OSCILLATIONS**

#### A. Theoretical Studies.

The problem of an isolated bubble of near spherical shape in a liquid of small viscosity has been treated in the framework of potential flow with a thin boundary layer near the bubble surface. An asymptotic method combining domain perturbation and multiple time-scale techniques was used to derive the governing equations and boundary conditions at successive orders of approximation. A slow time scale for the resonant interaction between radial and shape mode was defined, and dynamical equations were derived for the amplitude functions of the interacting oscillatory modes for resonant and near-resonant conditions. The specific cases of interest for this initial study were the two-to-one (and the one-to-one) resonance interactions where the frequency of the radial mode was twice (or equal to) that of an interacting high order shape mode.

In an earlier study 10 the stability of initially perturbed and radially oscillating bubbles in an *inviscid* liquid was analyzed via an examination of the solution trajectories for the radial mode and one nearly resonant shape mode in an equivalent planar representation. For the two-to-one case, it was shown that for a radial deformation exceeding a given threshold, a homoclinic orbit emanates from a fixed point corresponding to purely radial oscillations. This implies the possibility for producing chaotic dynamics upon the introduction of a time-varying pressure perturbation. For higher viscosity systems, the dynamics become more complicated  $2^{6}$ . It was established, however, that a continuous energy exchange takes place between the resonant radial and shape modes on a long time scale. Whenever two-to-one resonance conditions are achieved (higher oscillation amplitude is reached) this process is accelerated.

In a subsequent study <sup>27</sup>, the effect of viscosity was introduced in addition to time-dependent isotropic and non-isotropic (asymmetric) pressure perturbations. Bifurcation analysis of the amplitude equations for the two types of oscillatory modes reveals that for a sufficiently large amplitude in the volume mode, the onset of instability may lead to chaotic oscillations in both volume and shape modes. It was found, however, that a critical degree of detuning between the volume and shape resonance was required for chaos to occur. If an asymmetric pressure perturbation directly forces the shape oscillations, chaos was found to occur even for exact resonance; no detuning was required. Figure 3 reproduces a schematic description of the bifurcation sets, projected on the plane of the asymmetric drive amplitude ( $\Delta_n$ ) and the drive frequency detuning ( $\sigma$ ). Here  $\sigma$  is the nondimensional parameter expressing the difference between the frequency of the pressure perturbation and the frequency of the shape mode resonant with the volume mode. The regions with one fixed point correspond to stable oscillation of the coupled shape and volume modes at fixed amplitude. In regions with larger  $\sigma$ , the amplitude of the radial mode is typically smaller than the shape mode. However, if we traverse the diagram at a constant  $\Delta_n \sim 3$ , and decreasing values of  $\sigma$ , the needle-like regions correspond to a saddle-node bifurcation across which the solution changes to one with a much stronger radial mode. Within the needle, two steady solutions coexist - one with the amplitudes of the radial and shape modes much stronger than the other. Finally, within the parabolic region around  $\sigma = 0$ , the amplitudes of the shape and volume modes are time modulated -with the dynamics being either periodic on a slow time scale, or chaotic depending upon the specific values of  $\Delta_n$  and  $\sigma$ .

### B. Experimental Studies.

Usually, investigation of nonlinear bubble dynamics is carried out with ultrasonic methods consisting of trapping a single sub-critical bubble at a pressure antinode in a standing wave. As the bubble grows and reaches critical size, the carrier frequency of the standing wave matches the resonant volume mode frequency,

and the bubble oscillates at high amplitude. The pressure perturbations are therefore quasi-isotropic, and the volume mode is always driven first. On the other hand, the theoretical work described above, suggests that energy exchange can also take place in the reverse direction: from the shape to the volume mode. We have explored this alternate path to study the mode coupling problem, although we also intend to pursue the study of this problem via the direct excitation of the volume mode.

In order to satisfy the matching of the resonance conditions for centimeter-size bubbles, one must reduce the hydrostatic pressure in order to lower the volume mode resonance frequency to match a resonant shape mode of sufficiently low order to reduce viscous dissipation and to achieve significant oscillation amplitude. Using an apparatus described in references 11 and 12 placed in a vacuum chamber, we have been able to trap 0.5 cm diameter bubbles in 1 G at an ambient pressure down to 0.25 atmosphere. We have also verified that modulation of the ultrasonic standing wave allows the excitation of shape modes up to order 6 with *macroscopic* oscillation amplitude. The Earth's gravitational field prevents us from trapping a bubble of such a size at a lower ambient pressure due to the onset of cavitation in the host fluid.

The microgravity environment allows a significant relaxation in these experimental constraints by greatly reducing the necessary power to position a single bubble of 1 cm in diameter. The hydrostatic pressure can be further lowered, and a match between the volume mode frequency and twice the frequency of a shape mode of order 4 or 5 can be easily obtained. Thus the excitation of the fifth shape mode at a frequency of 138 Hz and at 0.045 atmosphere hydrostatic pressure should allow the observation of the radial mode excitation at 276 Hz when a 0.5 cm air bubble is trapped in water.

# IV. LARGE AMPLITUDE SHAPE OSCILLATIONS OF CHARGED AND UNCHARGED DROPS IN AN ELECTRIC FIELD

#### A. Theoretical Studies.

Using a spheroidal approximation for the equilibrium drop shape, an initial investigation of the dependence of drop oscillation frequencies on the presence of electric charges or of an electric field was carried out  $^{21}$ . Predictions of the resonant frequency changes due to mean drop deformation, of the effects of resonant coupling between time-periodic oscillations in the electric field and the drop shape, and of the conditions for transition to chaotic oscillations of the drop shape have been obtained for a conducting drop in a time-dependent electric field.

A more recent numerical treatment using the boundary element method  $^{28}$  has investigated the variations of the fundamental shape mode resonance frequency for charged and uncharged drops as function of the electric field strength and the drop charge. The results agree with those of Feng and Beard  $^{20}$  for small values of the electric field strength, but differ at the higher values. The variations of the resonance frequency on the amplitude of shape oscillations has also been calculated. Additional calculations involving the effects of a sinusoidal time-varying electric field was also obtained. Some of these results can readily be compared with the experimental data described below.

#### **B.** Experimental Studies.

The dynamics of ultrasonically levitated drops in the presence of a static and/or time varying electric field have been investigated in order to quantify the effect of a static field on the oscillation frequencies, the effects of a static shape distortion on the resonances of the droplet, the dynamic response of the drop to a sinusoidal time-varying E field, and the effect of charges on the drop dynamics. The advantage of a hybrid levitation system is the partial decoupling of the electric field and the levitation function, thus allowing the measurement of the influence of the E field in the absence of surface charges.

Figure 4 reproduces experimental data for the measurement of the resonance frequency of the fundamental mode of shape oscillation as a function of amplitude for *driven* oscillations. The decreasing trend is in agreement with the theoretical results described above. In this case, the oscillations are driven by a sinusoidal varying electric field. The drop is ultrasonically levitated, and has an oblate spheroidal equilibrium shape.

Figure 5 reproduces experimental data for the response of a drop to a frequency sweep in the increasing and decreasing frequency direction. Hysteresis is clearly present for significant oscillation amplitude.

# V. SUMMARY

Theoretical treatments of the resonant volume to shape oscillations mode coupling has provided strong evidence for the possibility of direct energy transfer under specific circumstances. The experimental effort has demonstrated the capability of exciting higher order shape modes at a reduced hydrostatic pressure, and parameters for a low gravity investigation have been delineated. Direct comparison between a numerical model and experimental results for the dynamics of levitated charged and uncharged drops in a static and time varying electric field has been possible. Hysteresis has been uncovered in the frequency response of droplets modulated by a sinusoidal varying electric field.

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Figure 1.

Comparison of ground-based results for the measurement of the bifurcation point for the equilibrium shape of rotating free drops. Measurement in 1G involves statically distorted drops while the conditions of microgravity allows experimenting with no shape distortion.









Comparison of ground-based and microgravity data for the resonant mode frequencies of a free bubble. The data are from P.L. Marston, E.H. Trinh, J. Depew, and TJ Asaki, IUTAM Symposium Proceedings (1993)

Experimental results of the measurement of the frequency shift as a function of the driving electric field amplitude. The drops are ultrasonically levitated and electrically shape modulated. The Weber number We is proportional to the square of the electric field magnitude.





Bifurcation sets in the frequency  $(\sigma)$  and perturbation amplitude  $(\Delta_n)$  space.

# Figure 5.

Experimental evidence for hysteresis in the frequency response of levitated drop driven by a sinusoidal timevarying electric field. The sense of the arrows shows the frequency sweep direction.