Multistability in the dynamics of a spherical cavitation bubble

Booklet of the Ph.D. Dissertation

Written by: Roxána Varga
MS.c. in Applied Mathematics

Supervisor: Dr. Ferenc Hegedűs
associate professor

Budapest, December 19, 2019
1. Introduction

Irradiating a liquid domain with high amplitude and high frequency ultrasound, small bubbles may be created inside the liquid. Due to the oscillating pressure field, the total pressure inside the bulk liquid becomes negative in the rarefaction phase of the sound wave [1]. As a consequence, the tensile stress caused by the acoustic wave gets extremely strong, stronger than the tensile strength of the liquid (assumed continuum). These effects lead to the formation of gas/vapor bubbles, which then oscillate radially and move in space as the pressure field varies in the liquid. In the rarefaction phase of the sound wave, they grow in size due to evaporation and expansion. Then, within just a fraction of a second, the negative pressure becomes overpressure, and the bubbles shrink in size or even rapidly collapse due to the compression phase. The collapse of the bubble can be so strong, that the pressure and the temperature at the collapse site can get as high as 1000 bar and 5000 K, respectively [2]. After the collapse, a spherically symmetric pressure wave is launched, which may evolve into a shock wave. Sonoluminescence, light emission is the other phenomenon associated with the collapse [3].

This phenomenon is called acoustic cavitation, and its physical effects led some scientists to study the phenomenon theoretically, and others to find possible ways to employ them in different applications.

1.1 Applications associated with acoustic cavitation

Acoustic cavitation and the effects associated with the collapse of the bubble plays a vital role in different fields of industry. One of the great success stories in modern chemistry to increase the chemical yield is the utilization of ultrasound, a technique called sonochemistry. For instance, one of the keen interests of sonochemistry is the generation of hydroxyl (’OH), hydrogen (H’), and perhydroxyl (HO’2) radicals and hydrogen peroxide (H2O2). The generation of the former two radicals usually happens inside the bubble when containing oxygen and water vapor due to the high temperature at the collapse [4]. In wastewater cleaning, for example, the generation of these highly reactive free radicals is essential for oxidation processes [5]. The use of ultrasound is a promising method also to clean the pores and the surface of fouled membranes or solid surfaces.

An essential quality of ultrasound enhanced technologies is that they are usually more cost-friendly compared to other commercial methods. Therefore, this research area is continually growing, searching for new ways of applications and optimization of the sonochemical reactors.

1.2 The aims of the thesis

On the topic of acoustically excited bubbles, several books, reviews, and numerical studies were written in the past few decades. The accumulated computational results have revealed the very complex nature of such a system, for instance, the structure of regular periodic and chaotic windows as a function of a single control parameter, or the resonance properties with respect to
1. INTRODUCTION

the driving frequency. These are all the consequences of the strongly nonlinear behavior of a single cavitation bubble under harmonic driving.

Another relevant property of nonlinear systems is that at a given parameter set, more than one stable periodic and/or chaotic solutions may coexist. From the application point of view, the multistability of the system usually has to be controlled as different stable solutions may mean significantly different system performance (e.g., the chemical output of a bubble) or risk the reproducibility and reliability. The basin of attraction of the different stable attractors usually form a fractal-like set in the state space; hence, the proper choice of the initial conditions are not always possible. Even if the initial condition is adequately chosen, some noise in the system could push the system to another attractor. Therefore, controlling of multistability is a highly researched topic in nonlinear dynamics. Over the past few years, several control techniques have been created.

Our group, led by my supervisor, in cooperation with the University of Göttingen (Drittes Physikalisches Institut, Georg-August-Universität Göttingen, the research group of Prof. Werner Lauterborn, Prof. Ulrich Parlitz and Dr. Robert Mettin) successfully developed a control method that can "select" any stable solution without feedback so that the system is temporarily operated by two frequency excitation [6]. The method works well for harmonically driven non-linear oscillators (like an acoustically driven single bubble) via a temporary addition of a second harmonic component to the driving. That is, the system has a temporary dual-frequency driving. One drawback of this method is that for proper control, the detailed bifurcation structure of the system is necessary. Therefore, the primary goal of the present dissertation is to describe the bifurcation structure of a single-frequency-driven single gas bubble and explore the multistable regions in the pressure amplitude-frequency parameter plane at different bubble sizes.

In order to support the above mentioned theoretical understanding of dual-frequency-driven single bubbles, dual-frequency measurements were carried out at the Georg-August University Göttingen, Germany, funded by the German Academic Exchange Service (DAAD - Deutscher Akademischer Austauschdienst) with a short term research scholarship. From the video recordings, the reconstruction of the radial oscillation of an individual bubble in the cluster was carried out with digital image processing techniques. We propose a numerical technique to reconstruct the properties (parameters) of the observed bubble from such data by comparing the recorded bubble radius-time curves with numerical simulations. The extracted amount of information and their validity is dependent on the complexity of the model itself. The main technical difficulty is the determination of the emerging unknown parameters by comparing the simulated and the measured bubble radii as functions of time via the definition of a suitable error function.

The numerical calculations were carried out using an in-house code written in C++/CUDA C to exploit the high computational capacities of GPUs and accelerate the identification of the unknown system parameters.
2. Bifurcation structure of the bubble oscillator

This chapter is dedicated to the exploration of the bifurcation structure of the bubble oscillator, namely the Keller–Miksis equation [7]. With a thorough parameter study in the parameter space of the harmonic driving, the bifurcation structure is revealed with the pressure amplitude \( p_A \) of the harmonic driving as a control parameter at different driving frequencies. We shall see that below the linear resonance frequency of the bubble, the bubble oscillator is mainly monostable, while above it, the multistability of the oscillator gets stronger as the frequency increases. Results revealed that the bubble oscillator shows a well-organized bifurcation structure in this two-parameter space.

2.1 The mathematical model

In order to model large amplitude, collapse-like oscillations of a single gas bubble in water, the Keller–Miksis equation is used:

\[
\left(1 - \frac{\dot{R}}{c_L}\right) R \ddot{R} + \left(1 - \frac{\dot{R}}{3 c_L}\right) \frac{3}{2} \dot{R}^2 = \frac{1}{\rho_L} \left(1 + \frac{\dot{R}}{c_L}\right) (p_L - p_\infty(t)) + \frac{R}{\rho_L c_L} \frac{d(p_L - p_\infty(t))}{dt}.
\]

This nonlinear, second-order ordinary differential equation describes the time evolution of the radius \( R(t) \) of a spherical bubble. Since it takes into account the compressibility of the liquid domain to the first order, it incorporates damping due to sound radiation. The dot stands for the derivative with respect to time. In Eq. (2.1), \( \rho_L \) and \( c_L \) are the density of the liquid and the speed of sound in the liquid, respectively.

The pressure far away from the bubble is

\[
p_\infty = P_\infty + p_A \sin(\omega t),
\]

where \( P_\infty \) is the static or ambient pressure, \( p_A \) is the amplitude of the pressure oscillation and \( \omega \) is the angular frequency of the periodic excitation of the ultrasonic irradiation. The mechanical equilibrium at the bubble wall can be written as

\[
p_G + p_V = p_L + \frac{2 \sigma}{R} + 4 \mu_L \frac{\dot{R}}{R},
\]

where the total pressure inside the bubble is the sum of the partial pressures of the noncondensable gas content \( p_G \) and the vapor pressure \( p_V \). The pressure at the bubble wall in the liquid side

\[
\frac{1}{\rho_L} \left(1 + \frac{\dot{R}}{c_L}\right) (p_L - p_\infty(t)) + \frac{R}{\rho_L c_L} \frac{d(p_L - p_\infty(t))}{dt}.
\]
is \( p_L \), \( \sigma \) is the surface tension and \( \mu_L \) is the dynamic liquid viscosity. The gas inside the bubble obeys a simple polytropic relationship:

\[
\begin{align*}
p_G &= \left( \frac{2\sigma}{R_E} - p_V + P_\infty \right) \left( \frac{R_E}{R} \right)^{3n}, \\
&\quad \quad (2.4)
\end{align*}
\]

where \( R_E = 0.1 \text{ mm} \) is the equilibrium radius (size of the bubble in stationary case) and \( n = 1.4 \) is the polytropic exponent.

For the numerical simulations, Eqs. (2.1)-(2.4) were rewritten into a system of first-order dimensionless differential equations. For this the dimensionless time \( \tau = t\omega/(2\pi) \), the dimensionless bubble radius \( y_1 = R/R_E \) and the bubble wall velocity \( y_2 = 2\pi R/(R_E\omega) \) were used.

Since the angular frequency can vary on a scale of many orders of magnitude, it is reasonable to normalize it with a suitable reference quantity. Therefore, a dimensionless relative frequency is used for the computations defined as

\[
\omega_f = \frac{\omega}{\omega_0},
\]

where \( \omega_0 \) is the linear resonant frequency of the system. According to [8], \( \omega_0 \) can be calculated from the following equation:

\[
\omega_0 = \sqrt{\frac{3n(P_\infty - p_V)}{\rho_L R_E^2}} + \frac{2(3n - 1)\sigma}{\rho_L R_E^3} - \frac{8\mu_L^2}{\rho_L^2 R_E^4}.
\]

The computations were carried out using clear water at constant ambient pressure \( P_\infty = 1 \text{ bar} \) and at constant ambient temperature \( T_\infty = 25^\circ C \). These ambient quantities specify all the liquid material properties determined by means of the Haar–Gallagher–Kell equation of state [9].

### 2.2 The bifurcation structure

To find the stable orbits at given parameters, the most commonly used method is to take an initial value problem (IVP) solver and simulate the transient solution through as many cycles as it takes for the convergence to a stable solution (attractor). After the convergence, the different properties of a solution can be saved, such as the Poincaré section \( P(y_1) \) and \( P(y_2) \), and the period \( m \). The basic means to explore the bifurcation structure of the attractors are pressure amplitude response curves, where the first coordinate of the Poincaré plane \( P(y_1) \) is plotted versus the control parameter \( p_A \) at different relative frequency \( \omega_f \). In order to find all the relevant stable orbits, at each parameter set, 5 IVPs were solved with randomly chosen initial conditions. In both diagrams in Fig. 2.1, the pressure amplitude \( p_A \) was varied between 1 bar and 5 bar with an increment of 0.01 bar.

Panel A) shows the bifurcation structure below the linear resonance frequency \( (\omega_f = 0.5) \). Here the initial period-1 orbit, having emerged from the dimensionless equilibrium radius \( y_{1,E} = 1 \) of the unexcited system, undergoes a period-doubling (PD) bifurcation near a pressure amplitude of \( p_A = 1 \text{ bar} \) and becomes unstable. Here, a narrow band of chaotic solutions also exists, indicated by the scattered Poincaré points. The upper branch of the period-1 curve, appearing via a saddle-node (SN) bifurcation, exhibits a Feigenbaum PD cascade transforming gradually into a large chaotic domain.

At \( \omega_f = 2 \) relative frequency in panel B), the initial period-1 solution undergoes a \( PD \) bifurcation at very low pressure amplitude \( p_A \). This is a well-known characteristic property of
2.2. THE BIFURCATION STRUCTURE

Figure 2.1: Pressure amplitude response curves of the dimensionless bubble radius at the different relative frequencies: A) $\omega_f = 0.5$ and B) $\omega_f = 3$.

nonlinear systems excited at the frequency value two times their resonance frequency [10, 11]. The emerging period-2 region exists approximately up to a pressure amplitude of $p_A = 3$ bar. Meanwhile, a co-existing period-3 orbit appears via a $SN$ bifurcation and finishes in a $PD$ cascade. Above $p_A = 3.5$ bar, the bifurcation structure alternates between periodic and chaotic windows.

Figure 2.2: Codimension-two bifurcation curves corresponding to subharmonic resonances of order $(1,m)$, where the period $m$ is between 1 and 9, and to their subsequent period-doubling points. The solid and dashed lines are the saddle-node $SN$ and the period-doubling $PD$ curves, respectively.

From the multistability point of view, below the linear resonance frequency of the bubble, the system is mainly monostable. Even though at $\omega_f = 0.5$ period-2 and period-3 stable solutions coexist with the period-1 solution, their range is so small, that the control of multistability has
2. BIFURCATION STRUCTURE OF THE BUBBLE OSCILLATOR

minor importance. However, with increasing $\omega_f$, the multistability of the system dominates the majority of the parameter ranges, making the bifurcation structure of the oscillator extremely complex.

For the description of the bifurcation structure of the bubble oscillator Eq. (2.1), the torsion number $n$ and the period $m$ of a stable attractor are used. The torsion number of a periodic attractor is the average number of rotations of neighboring trajectories around the closed orbit during one of its periods; it is an integer number near a bifurcation point. According to [12], the torsion number $n$ and the period $m$ together are an efficient tool to label the resonances of nonlinear oscillators.

To obtain a global overview of the subharmonic resonances in the excitation parameter plane, the $SN$ bifurcation points of the bifurcation structure at $\omega_f = 5$ and their first $PD$ points in the Feigenbaum cascade are tracked down as a function of the pressure amplitude $p_A$ and relative frequency $\omega_f$. Here only the subharmonic resonances are computed; these are the bifurcation curves of order $(1, m)$. This task can be easily done by reformulating the problem into a boundary value problem (BVP). The resulting codimension-two bifurcation curves are summarized in Fig. 2.2. The solid and the dashed curves are related to the $SN$ and $PD$ bifurcation points, respectively. Here, a remarkable U-shaped ordering of the subharmonic structure arises, which was hidden in the one-dimensional diagrams shown in Fig. 2.1.

**Thesis #1**

Let us consider the dimensionless form of the Keller–Miksis bubble oscillator with a simple harmonic driving $p_A \sin(\omega t)$. Let the liquid be water, and its parameters are calculated from the Haar–Gallagher–Kell equation of state with ambient temperature $T_\infty = 25^\circ C$ and ambient pressure $P_\infty = 1$ bar. Let us consider, in the harmonic driving, the dimensionless relative frequency $\omega_f = \omega / \omega_0$, where $\omega_0$ is the linear resonance frequency of the system, and the equilibrium bubble radius $R_E = 0.1$ mm.

Then, above the linear resonance frequency, the subharmonic resonances of the bubble oscillator, in the parameter field of the driving parameters $p_A$ and $\omega_f$, form a U-shaped structure. The order of these curves is $(1, m)$, where 1 is the torsion number, and $m = 1, 2 \ldots$ is the period. The structure of these bifurcation curves can be described with a one-sided Farey tree, as depicted in Fig. 2.3. Furthermore, the bifurcation structure is strongly multistable above $\omega_f = 2$ relative frequency.

\[\text{Figure 2.3: Structure of the subharmonic resonances described via a one-sided Farey tree.}\]

Related publications: [T1], [T5]
3. The role of equilibrium bubble size in bubble dynamics

Usually, in a bubble cloud, the size of the bubbles is not uniform. It is typically distributed in the range of 1 \( \mu \text{m} \) to 0.1 mm, and in water, the largest bubble size is about 0.1 mm. Therefore, from the applications point of view, it is important to take into account the size of the bubbles.

Previously, Behnia and his co-workers, using the Keller–Miksis equation, found that the bifurcation structures of different pressure amplitude response curves are remarkably similar when \( R_E \omega \) is kept constant, but detailed physical explanation and range of validity was omitted [13]. Here \( R_E \) is the equilibrium radius (size of the unexcited bubble), and \( \omega \) is the excitation angular frequency of the periodic driving. In this chapter, this problem is revisited using the same equation, the Keller–Miksis oscillator, as applied by Behnia et al. [13]. The detailed analytical and numerical investigations reveal the range of the applicability of the condition of Behnia et al., which is affected by the viscosity and the surface tension. In the case of water, the influence of the viscosity is negligible, while the effect of surface tension becomes important at bubble size lower than 5 \( \mu \text{m} \).

3.1 Similarities of the bifurcation structures

To obtain a quick overview of the bifurcation structure of the bubble oscillator, two-dimensional bi-parametric plots (contour plots) are generated with the equilibrium bubble size and the pressure amplitude as control parameters. The number of the applied equilibrium radii \( R_E \) is 991, distributed linearly between 1 \( \mu \text{m} \) and 0.1 mm. The pressure amplitude \( p_A \) was varied between 1 bar and 5 bar with an increment of 0.01 bar. Here, at each \( p_A - R_E \) parameter pair, 10 IVPs are solved with randomly chosen initial conditions. To show the evolution of the bifurcation structure efficiently on the \( p_A - R_E \) planes, the color-coded period of the stable solutions are presented.

In Fig. 3.1 two bi-parametric plots are shown at \( \omega_f = 0.5 \) and \( \omega_f = 2 \) relative frequencies. Instead of the first component of the Poincaré section \( P(y_1) \), the color-coded period of the found attractors is presented to be able to trace the bifurcation points easily. Here solutions with period-8 or higher—including chaotic oscillations—are colored black. In both the cases of \( \omega_f = 0.5 \) and \( \omega_f = 2 \), above about 4.5 bar and about 3.3 bar pressure amplitude, respectively, the black bands imply that the largest period of the found attractors is higher than 8.

The results in the \( p_A - R_E \) planes show clearly that either below or above the linear resonance frequency of the bubble, there is no sharp threshold (in terms of the bubble size \( R_E \)) for the uniformity of the structure. The bifurcation points are slowly moved towards higher pressure amplitudes with a decreasing equilibrium bubble radius. The chaotic regime disappears for very small equilibrium bubble radii in the investigated pressure amplitude region. The red lines in Fig. 3.1 the chosen threshold value \( R_E = 5 \mu \text{m} \) is highlighted by the red lines. For bubble sizes
3. THE ROLE OF EQUILIBRIUM BUBBLE SIZE IN BUBBLE DYNAMICS

Figure 3.1: Bi-parametric bifurcation structure of the bubble oscillator at relative frequencies $\omega_f = 0.5$ (left) and $\omega_f = 2$ (right). The colorbar represents the period of the found attractors up to period-7 in the $p_A - R_E$ plane. Solutions with period-8 or higher are colored black.

above this threshold, the bifurcation structures with respect to the pressure amplitude are nearly identical.

In the following, the condition of Behnia is examined via dimensional analysis of the dimensionless equation system. Results show that there are only two kinds of terms that still have an explicit dependence on $R_E$ when $C = \frac{R_E \omega}{2 \pi}$ is constant, namely,

$$\frac{2\sigma}{R_E}, \quad \text{and} \quad \frac{4\mu_L}{\rho_L R_E}.$$ (3.1)

It follows that when the terms containing the dynamic viscosity of the liquid $\mu_L$ and the surface tension $\sigma$ are negligible, the bifurcation structure does not change when $C = \frac{R_E \omega}{2 \pi}$ is kept constant. This gives a precise explanation of why the bifurcation structures are similar in a wide range of parameters in the study of Behnia et al. [13]. Yet, the influences of $\sigma$ and $\mu_L$ on the bifurcation structure with respect to the equilibrium bubble radius $R_E$ have to be still clarified.

3.2 The linear resonance frequency of the bubble

We determine the relationship between the conditions of Behnia

$$R_E \omega = \text{constant} \quad \text{(3.2)}$$

and the present study

$$\omega_f = \frac{\omega}{\omega_0} = \text{constant}. \quad \text{(3.3)}$$

The linear resonance frequency $\omega_0$ of the system, Eq. (2.6), multiplied by $R_E$ is

$$\omega_0 R_E = \sqrt{\frac{3n(P_\infty - p_v)}{\rho_L} + \frac{2(3n - 1)\sigma}{\rho_L R_E} - \frac{8\mu^2_L}{\rho^2_L R^2_E}}, \quad \text{(3.4)}$$
which can be further simplified to

$$\omega_0 R_E = \sqrt{\frac{3n(P_\infty - p_v)}{\rho_L}} \approx \sqrt{\frac{3nP_\infty}{\rho_L}}$$  \hspace{1cm} (3.5)$$

if the effects of the surface tension $\sigma$ and the liquid dynamic viscosity $\mu_L$ are neglected. Observe that these assumptions are exactly the same as in the case of the condition of Behnia in Eq. (3.2). Observe also that in Eq. (3.4) exactly the same terms appear as presented in Eqs. (3.1). Now, it is clear that if $R_E \omega$ is constant, then the relative frequency

$$\omega_f = \frac{\omega}{\omega_0} = \frac{R_E \omega}{R_E \omega_0}$$  \hspace{1cm} (3.6)$$
is constant as well, implying that conditions Eq. (3.2) and Eq. (3.3) are identical when $\sigma, \mu \to 0$.

![Figure 3.2](image)

**Figure 3.2:** The linear resonance frequency $\omega_0$ of the bubble as the function of the equilibrium radius $R_E$. When both the damping $\mu_L$ and the surface tension $\sigma$ are taken into account, its evolution is shown by the black line. The undamped eigenfrequency is shown by the blue line, while the red line shows when both the effects of $\mu_L$ and $\sigma$ is neglected.

The validity range of these two conditions is examined through the analysis of the linear resonance frequency Eq. (2.6). Figure 3.2 shows how the two terms of Eqs. (3.1) effect $\omega_0$ in a
wide range of equilibrium radius on a logarithmic scale. The black line is the full equation, the blue line represents the case when $\mu_L$ is neglected, and the red line shows when both $\mu_L$ and $\sigma$ are zero.

Examining the 1% difference between the colored lines and the black line, a threshold can be established for the bubble radius. For equilibrium radii larger than about $R_E = 0.05 \mu m$, there is no significant difference between the black and the blue line, which means that in this range of the bubble size, the effect of $\mu_L$ is negligible, see Fig. 3.2B. When both terms related to $\sigma$ and $\mu_L$ are neglected (solid red line), the linear resonance frequency starts to differ from the black line by 1% for equilibrium radii smaller than about $R_E = 5 \mu m$. Since the effect of the liquid (water) dynamic viscosity is not significant for bubble sizes larger than $R_E = 0.05 \mu m$, here, the difference between the black and the red lines is caused solely by the surface tension. These findings are in very good agreement with the results obtained by analyzing the bifurcation structure itself (see again Fig. 3.1).

**Thesis #2**

Let us consider the Keller–Miksis bubble oscillator with a simple harmonic driving $p_A \sin(\omega t)$. Let the liquid be water, and its parameters are calculated from the Haar–Gallagher–Kell equation of state with ambient temperature $T_\infty = 25^\circ C$ and ambient temperature $P_\infty = 1$ bar. Let us consider the dimensionless relative frequency $\omega_f = \omega / \omega_0$, where $\omega_0$ is the linear resonance frequency of the system.

The bifurcation structure of the dimensionless form of the oscillator depends on two terms, when $R_E \omega/2\pi$ is constant, namely:

$$\frac{2\sigma}{R_E}$$

and

$$\frac{4\mu_L}{\rho_L R_E},$$

where $\sigma$ is the surface tension, $\mu_L$ is the liquid dynamic viscosity and $\rho_L$ is the liquid density.

Based on the linear resonance frequency $\omega$ of the system, the term containing $\mu_L$ can be neglected for equilibrium radii larger than about $R_E = 0.05 \mu m$, and $\sigma$ is not significant for bubble sizes larger than $R_E = 5 \mu m$. For bubbles smaller than these conditions, the bifurcation structure shifts to higher pressure amplitudes without qualitative change.

Related Publications: [T2], [T6], [T7]
4. The role of liquid dynamical viscosity in bubble dynamics

Previously we saw that the liquid dynamic viscosity $\mu_L$ and the surface tension $\sigma$ play an important role in the oscillation for bubbles smaller than a threshold: the bifurcation structure shifts to higher pressure amplitudes. In this chapter, we are going to explore the inner structure of the period-3 subharmonic resonance in the 2-dimensional parameter space of the excitation and investigate the effect of the liquid dynamic viscosity on it.

We must mention that previously Klapcsik et al. [14] successfully explored the complete family of the period-3 subharmonic resonances of their model, which was also the Keller–Miksis bubble oscillator, and found that it forms a zig-zag pattern in the pressure amplitude-frequency parameter plane of the external forcing. They found that this structure is organized by a special two-dimensional isoperiodic structure, called shrimp-shaped domains (SSD). The special property of their system was the very strong dissipation rate that originated from the high viscosity of the liquid domain (glycerin). Interestingly, in our results obtained in water having orders of magnitude lower damping rate, the above-mentioned SSDs are absent. Consequently, the dissipation rate must play a significant role in the formation of SSDs, at least in case of the Keller–Miksis nonlinear oscillator.

4.1 The effect of viscosity on the internal structure of the period-3 subharmonic resonance

To reveal the possible routes to the shrimp-shaped domains via the formation of the zig-zag pattern, the subharmonic resonance family of the period-3 solution was computed at different dynamic viscosities $\mu_L$, from which two are presented in Fig. 4.1. The top panel and bottom panels show the cases of low and high dynamical viscosities, respectively. With increasing dynamic viscosity $\mu_L$, two main alterations in the bifurcation structure can be recognized. First, all the curves are shifted towards the higher pressure amplitudes. Second, the block patterns $a)$ and $b)$ gradually approach to and collide with another pair of bifurcation curves initiating a complex interaction between two $SN – PD$ pairs. As the viscosity increases, this phenomenon takes place first with block patterns presented at high pressure amplitudes and gradually spreads towards the lower amplitude regions forming the zig-zag pattern already observed in [14].

Here we only show the results of the computations for block pattern $a)$ in Fig. 4.2, which shows the magnified examples of building block $a)$ in the zig-zag pattern before and after the interaction of their $SN – PD$ bifurcation curve pairs. Figure 4.2A is the magnification of a block pattern $a)$ at $\mu_L = 0.100 \text{ Pa.s}$. At this viscosity value, these $SN – PD$ pairs are not connected; that is, this state is without the interaction of the bifurcation curves. Figure 4.2B and shows the same bifurcation curves as panel A) does but at an increased dynamic viscosity value at $\mu_L = 0.252 \text{ Pa.s}$. Here the $SN$ bifurcation curves have exchanged their subsequent $PD$
4. THE ROLE OF LIQUID DYNAMICAL VISCOSITY IN BUBBLE DYNAMICS

Figure 4.1: Top panel: The internal structure of the subharmonic resonance family of order $SN(1, 3)$ (solid purple line, see also 2.2). The arrows show the type and order of the first few bifurcation curves. Bottom panel: The same internal structure of the subharmonic resonance family of order $SN(1, 3)$ at orders of magnitude higher viscosity. The zig-zag pattern of the bifurcation structure is clearly visible in this case.

In order to find proof that in Fig. 4.2B, a shrimp-shaped domains was indeed created, IVP scans are also provided in the right column of the figures. These diagrams show the color-coded period (up to period-7) of the stable periodic solutions found. Chaotic solutions are again omitted in order to find the periodic structures possibly hidden by them. The period-3 and period-6 solutions are colored light pink and gray, respectively. Color white means that...
4.1. THE EFFECT OF VISCOSITY ON THE INTERNAL STRUCTURE OF THE PERIOD-3 SUBHARMONIC RESONANCE

Figure 4.2: Resonance curves of block pattern a) creating a shrimp-shaped region as the liquid dynamic viscosity increases from $\mu_L = 0.100$ Pa s (top row) to $\mu_L = 0.252$ Pa s (bottom row). Panel A) and B) are the magnifications of the black rectangles in Fig. 4.1A) and B), respectively. Panel C) and D) are the results of an IVP scan of the parameter areas of panel A) and B), respectively. The color code means the period of the converged periodic solutions up to period-7. Chaotic solutions are omitted from these panels.

only chaotic solutions were found. The $SN$ and $PD$ bifurcation curves from panels A) and B) are also depicted in panels C) and D), respectively. These phase diagrams show that after the interaction of the bifurcation curves, an isoperiodic domain with period-3 is created inside the domain, bounded by the outer $SN$ and the two crossing $PD$ curves.

Thesis #3

Let us consider the dimensionless form of the Keller–Miksis bubble oscillator with a simple harmonic driving $p_A \sin(\omega t)$. Let the liquid be water, and its parameters are calculated from the Haar–Gallagher–Kell equation of state with ambient temperature $T_\infty = 25^0C$ and ambient pressure $P_\infty = 1$ bar. Let us consider the dimensionless relative excitation frequency $\omega_f = \omega/\omega_0$, where $\omega_0$ is the linear resonance frequency of the system, and the equilibrium bubble radius $R_E = 0.1$ mm.

In the two-dimensional parameter space of the external forcing $p_A$ and $\omega_f$, the internal structure of the period-3 solution with order $(1, 3)$ is mainly built up by pairs of saddle-node ($SN$) and period-doubling ($PD$) bifurcation curves. The formation of shrimp-shaped domains is highly dependent on the damping of the system. With the parameters determined for pure water (low viscosity), the two-dimensional bifurcation structure completely lacks these structures. Increasing the damping parameter (dynamic viscosity) of the system, two mechanisms for the creation of shrimp-shaped domains are present. In the first case, two $SN$ curves of the same order collide and interchange one of their branches, while in the other case, two $PD$ curves again with the same orders collide and form another pair of $PD$ bifurcations.

Related Publications: [T3]
5. Dual-frequency-excited bubble measurements

Recently, the attention has turned towards dual- or multi-frequency-driven systems due to their several positive effects [15]. However, the influence of another frequency on the oscillating bubbles or bubble clusters is still not well understood in terms of bubble dynamics. This prevents a coherent interpretation of the partly contradictory experimental results in the literature [16], as well as further optimization of dual-frequency driven sonochemical reactors.

As it was mentioned in the previous chapters, our group has made a significant step toward the theoretical understanding of dual-frequency driven single bubbles by employing a graphics processing unit (GPU) during the simulations [6]. This allows the investigation of much larger parameter space than it was possible with conventional approaches before (using CPUs). The main aim of the present chapter is to perform experiments to support the numerical simulation and propose a technique to identify the underlying model parameters of an individual oscillating bubble.

5.1 Dynamics of individual bubbles under dual-frequency excitation

During the measurements, I observed the ultrasound-induced bubbles with a high-speed camera with an exposure time of 1 µs at 162750 frames per second. In order to extract the radius of individual bubbles, digital processing was applied using the ImageJ software. In this study, only the oscillation of one bubble is presented. Figure 5.1 shows the resulting bubble radius curve as the function of time.

![Figure 5.1: The measured bubble radius time curve (solid black line). The maxima at every excitation period (determined by \( f_1 \)) are connected by the blue solid line.](image)

In every period, six or seven data points are obtained. In typical nonlinear bubble oscillation, the dynamics near the minimum radius have a concise time scale (fast bubble wall), whereas,
near the maximum, it has a relatively long time scale (slow bubble wall). Due to this effect and
the resolution limit, the smallest bubble radii along an oscillation period cannot be determined
precisely (or even cannot be observed at all). However, the periodic structure of the oscillation
responding to the maximum radii at each period is still well reconstructed. Therefore, the
comparison of this measured signal with numerical simulations is carried out by taking into
account only the respective “period maxima”.

5.2 The bubble model and the numerical method

For the computations, the Keller–Miksis bubble oscillator is used, defined by Eq. (2.1). In
order to consider dual-frequency sound radiation, the pressure far away from the bubble $p_\infty$ is
defined as

$$p_\infty = P_\infty + p_{A1} \sin(\omega_1 t) + p_{A2} \sin(\omega_2 t + \theta),$$

where $p_{A_i}$ and $\omega_i (i = 1, 2)$ are the amplitudes and frequencies of the excitations, respectively.
Their phase difference is denoted by $\theta$.

For the numerical computations, the angular frequencies of the excitation were set according
to the experiment: $\omega_1 = 2\pi \cdot 25000 \text{ rad/s}$ and $\omega_2 = 2\pi \cdot 50000 \text{ rad/s}$. To find the optimum
parameter set corresponding to the best fit of the calculated radius-time curves to the measured
data shown in Fig. 5.1, several simulations were computed with different parameter combina-
tions. The unknown parameter space includes the two pressure amplitudes $p_{A1}$ and $p_{A2}$, the
phase shift between the two sine waves $\theta$ and the equilibrium radius of the bubble $R_E$. Hence,
a 4D parameter scan was performed.

For the comparison of the simulated and the measured signal, first, the maximum of the sim-
ulated signal was sampled in every excitation period. Then it was compared with the measured
signal using cross-correlation to ensure the possible best starting point in time for further exami-
nation.

5.3 Results

As an initial attempt, three error functions and their weighted combination are defined as

$$\Delta_{\text{max}} = |\text{MAX}\{R_m\} - \text{MAX}\{R_s\}|,$$
$$\Delta_{\text{min}} = |\text{MIN}\{R_m\} - \text{MIN}\{R_s\}|,$$
$$\Delta_{\text{avg}} = |\text{AVG}\{R_m\} - \text{AVG}\{R_s\}|,$$
$$\Delta_w = \alpha_{\text{min}} \Delta_{\text{max}} + \alpha_{\text{max}} \Delta_{\text{min}} + \alpha_{\text{avg}} \Delta_{\text{avg}}$$

where $\Delta_{\text{max}}$, $\Delta_{\text{min}}$ and $\Delta_{\text{avg}}$ try to minimize the difference between the measured and the
simulated largest, smallest and averaged local maxima, respectively. $\Delta_w$ combines Eqs. (5.2)-(5.4) where the weights always chosen so that $\alpha_{\text{min}} = \alpha_{\text{max}}$ and $\alpha_{\text{avg}} = 1 - \alpha_{\text{min}} - \alpha_{\text{max}}$. For the
first three error functions, the optimum parameter set from the 4D scanned space is determined
and examined. Without showing here the results we found, that neither of the functions can give
an acceptable fit to the measurement. The inspection of the weighted error function in a wide
range of weight combinations showed that the global minima all the unknown parameters are
not changing when the weight $\alpha_{\text{min}}$ is not too small nor too big (see Fig. 5.2 for the case of $p_{A1}$).

However, considering the 10 and 20 smallest values of the error-functions, we concluded that
the ranges are the trivial parameter sets: the computational domain of the respected parameter.
Therefore the estimations are all loaded with a significant deviation.

Naturally, the best solution that fits the measured signal would be at a parameter set, where
all the four equations are minimal. The results presented in the dissertation proves that there is
a broad set of parameter combinations that almost equally well represent a good fit in terms of the corresponding error function. This is the primary difficulty of the present situation. In these situations, when the error function does not have a single optimum, but a large set, a traditional optimum search algorithm would not find all of them. Therefore, in our case, the used brute force technique is essential.

Unfortunately, this investigation proves that using only the maxima of the bubble radius time curve is not adequate for the numerical fitting of the Keller–Miksis equation. Such a large number of unknown parameters requires more than one quantity of the oscillation to fit a strongly nonlinear equation on the data. For example, knowing both the minima and the maxima of the oscillation would provide a limitation on the possible optima of a well-chosen error function. Alternatively, knowing both the equilibrium bubble size and the maxima of the oscillation would result in smaller unknown parameter space.

Generally, the curve fitting would be easier if the measurement points were denser in time. However, even with a fast camera, it is not possible to observe a free oscillating bubble for a long period of time with a large frame rate, because the bubble moves in space as well. Therefore, a proper numerical method, in our case, an error function is needed. A better error function could be one that takes into account the other measurement points as well, not only the local maxima. For this, a possible way is to compare the measurement data and the simulation by their re-sorted time series curves according to the period of the driving. An error function defined based on the re-sorted data points could result in a better estimate of all the unknown parameters. The importance of this comparison lies in the chaotic-like motion of oscillation: an exact match could never be found, but a statistically good enough one. The definition of such an error function is not part of this dissertation.

**Thesis #4**

The parameters needed to identify the underlying Keller–Miksis model of the dual-frequency excited bubble for the time signal measured with a high-speed camera are the pressure amplitudes of the excitation, the phase difference between the two driving signals, and the equilibrium size of the driven bubble. An error function based on the weighted sum of the average, minimum, and maximum values of the measured and calculated maximum bubble radius in every acoustic cycle is not feasible. Although the maxima of the bubble radii can be measured most accurately, an error function must include all the sampled points during a measurement.

Related Publications: [T4], [T8]
Own publications

Journal papers


Conference papers


Bibliography


