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Oscillations of soap bubbles

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Abstract. Oscillations of droplets or bubbles of a confined fluid in a fluid environment are found in various situations in everyday life, in technological processing and in natural phenomena on different length scales. Air bubbles in liquids or liquid droplets in air are well-known examples. Soap bubbles represent a particularly simple, beautiful and attractive system to study the dynamics of a closed gas volume embedded in the same or a different gas. Their dynamics is governed by the densities and viscosities of the gases and by the film tension. Dynamic equations describing their oscillations under simplifying assumptions have been well known since the beginning of the 20th century. Both analytical description and numerical modeling have made considerable progress since then, but quantitative experiments have been lacking so far. On the other hand, a soap bubble represents an easily manageable paradigm for the study of oscillations of fluid spheres. We use a technique to create axisymmetric initial non-equilibrium states, and we observe damped oscillations into equilibrium by means of a fast video camera. Symmetries of the oscillations, frequencies and damping rates of the eigenmodes as well as the coupling of modes are analyzed. They are compared to analytical models from the literature and to numerical calculations from the literature and this work.

S Online supplementary data available from stacks.iop.org/NJP/12/073031/ mmedia

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1. Introduction

In nature, one can find various types of confined fluids exhibiting surface oscillations; among them are bouncing droplets, fusing vesicles, rising bubbles in a liquid (e.g. in champagne or soda) or liquid drops evaporating on a heated plate (the Leidenfrost phenomenon). Recent interest in the dynamics of similar systems has been focused, for example, on the properties of bouncing drops [1]–[4], pendant and sessile drops [5]–[9], Leidenfrost droplets floating on a plane [10] or moving on structured surfaces [11, 12] and the motion of droplets on vibrating surfaces [13]–[18].

Generally, each of these examples involves two fluids, one enclosed by the other. Each fluid can be a liquid or gas. With respect to these two fluids, two limiting cases can be classified: if the enclosed fluid provides the dominant kinetic energy contribution and the outer fluid is negligible (density of the outer medium $\rho_0 \approx 0$), this structure is conventionally referred to as a drop. If the contribution of the inner fluid to the kinetic energy is negligible (density $\rho_i \approx 0$), the system is usually referred to as a bubble. We will use this notation here in the discussion of the literature data. In the general case, both fluids may contribute to the dynamics. The special situation where both fluids are of the same importance ($\rho_0 \approx \rho_i$) is considered here: in soap bubbles, there is gas on both sides of an interface made of a soap solution.

In the absence of external forces such as gravity, electric or magnetic fields, the equilibrium shape of the confined volume is a sphere for isotropic fluids. If the interface is not in equilibrium, shape transformations towards equilibrium are driven by the surface or interface tensions. In some cases, bending rigidity or spontaneous curvature of a membrane separating the fluids may be involved. The route towards equilibrium will normally have the form of (weakly) damped oscillations, under a permanent conversion of potential and kinetic energies. On the other hand, one can also excite the droplets (or bubbles) to oscillations about the equilibrium shape by continuous excitation with sound or other external forces. In this work, we study the relaxation of an initial non-equilibrium shape towards the minimum interface equilibrium state. The linear equations for oscillations of small amplitudes were already derived a century ago [19]–[21], and subsequently, several studies have dealt with the nonlinear treatment of the problem, considering e.g. mode couplings, frequency shifts with oscillation amplitude and viscous effects [22]–[30].

For the bubble sizes studied here, the properties of the thin liquid film (except for its surface tension) are irrelevant for the eigenmodes. The oscillation frequencies are comparatively slow

and weakly damped. This provides the experimental opportunity for a precise observation and recording of the oscillations even with moderate time resolution of the detector. A quantitative characterization of the dynamics is possible. All this, along with the simple preparation and observation of these objects, qualifies soap bubbles as an excellent model system to test predictions of theoretical models on droplet and bubble dynamics. But interestingly, quantitative experimental data on soap bubble oscillations are not available in the literature so far. The lack of quantitative experimental study of soap bubbles, as a model system with similar fluids on both sides of a thin liquid membrane.

A few peculiarities have to be considered: since both fluids are gases, we will have to discuss the incompressibility of the bubbles. As both inner and outer media have equal densities, an oscillation of the center of mass of the inner gas volume is detectable unlike in most other experimental studies in similar systems. Note however that in pendant (or sessile) drops this center of mass may undergo oscillations [5] because it can exchange momentum with the solid support.

The aim of the experiments reported here is to evaluate predictions of different models, put forward during the last century, and to test the validity of several assumptions. In order to trace reasons for possible discrepancies of experimental and theoretical results, we also compare our experimental data with the results of a numerical simulation of the full dynamic equations by a finite element method.

In order to keep the experiment simple, we have developed a special excitation technique that allows us to excite non-equilibrium states of high symmetry. The initial state is created by the fusion of two spherical soap bubbles floating in air. The axis connecting the two centers of the fusing bubbles remains, after coalescence, a symmetry axis of the system, and the oscillating bubbles retain this symmetry. This facilitates the analysis of the excited oscillation modes.

The paper is organized as follows: in section 2, we summarize the results obtained for oscillating bubbles and droplets in the literature. Then, the experimental setup and data evaluation are described in section 3. In section 4 we present experimental data obtained with representative soap bubbles, and in section 5, the results of a numerical simulation are introduced. The last section discusses and summarizes the findings.

2. Theoretical background

The problem of oscillating fluids was first considered by Kelvin [19] and Rayleigh [20] who calculated the special case of a drop. Lamb treated the more general case of two fluids, in which one of them is enclosed by the other one, in his book *Hydrodynamics* [21]. He simplified the system by considering only irrotational flow, non-viscous and incompressible fluids and only small, axisymmetric deformations. In his linear analysis, he identified the eigenmodes of the system to be the spherical harmonics Y_{ℓ}^m characterized by the mode number ℓ . The second index *m* is set to zero here because of the assumed axial symmetry of the oscillations, and we drop this index in the following. Hence the interface between both fluids may be expanded in a series:

$$r(t;\theta) = \sum_{\ell=0}^{\infty} A_{\ell}(t) Y_{\ell}(\theta),$$
(1)

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Figure 1. (a) Definition of the coordinates, (b) experimental setup for the observation of the oscillations of coalescing soap bubbles and (c) approach, attachment and fusion of two bubbles.

where θ is the polar angle and *r* is the distance between the coordinate center and the interface, measured in the direction θ (figure 1(a)). This is only applicable if the surface function $r(\theta)$ is unique, which holds for all experiments presented hereafter. Lamb derived the natural frequencies of these eigenmodes, related to the equilibrium radius *R* (of a sphere with the same volume as the deformed structure), the surface tension of the interface σ , the densities of the inner and outer fluids ϱ_i and ϱ_o and the mode number ℓ :

$$(\omega_{\ell})^{2} = \frac{\sigma}{R^{3}} \frac{(\ell-1)\ell(\ell+1)(\ell+2)}{\varrho_{i}(\ell+1) + \varrho_{o}\ell}.$$
(2)

The frequencies increase with the mode number. It must be pointed out that there are no natural frequencies of the zeroth mode and of the first mode. This is obvious because a decoupled oscillation of the zeroth mode, which is equal to a variation of the radius of the inner fluid, changes the volume and therefore violates the incompressibility condition, given by

$$V = 2\pi \int_0^\pi \int_0^{r(t;\theta)} r^2 \sin\theta \, d\theta \, dr = \text{const.}$$
(3)

When the small amplitude approximation does not hold, this condition leads to an oscillation of the zeroth mode, as will be discussed in section 4 in more detail. An oscillation of the first mode is not permitted because the center of mass must not oscillate unless an external force acts on the droplet (or bubble), it may only move in space at constant speed. In the linear approximation, the oscillation of the first mode of a drop with constant radius would simply mean a translation of the center of mass. Since there is no restriction as to where to place the origin of the coordinate system for the expansion in (1), it can always be chosen in the manner that the amplitude of the first mode is zero at all times, as was already pointed out by Becker *et al* [27]. Shaw [31] showed that for moderate amplitudes the oscillation of certain modes changes the center of mass and forces the inner liquid to translate along its symmetry axis. Every excited mode will induce such a motion.

Table 1. Coefficients $\omega_{\ell}^{(2)}$ for different mode numbers in the special cases of drops $\varrho_i \gg \varrho_o$ and bubbles $\varrho_o \gg \varrho_i$.

$\ell = 2$		$\ell = 3$		$\ell = 4$		
Drop Bubble		Drop Bubble		Drop	Bubble	
1.278 (1.170)	1.454	2.090 (1.998)	2.220	2.914 (2.797)	2.898	

Lamb [21] also estimated the damping constant for the special case of a drop, assuming small amplitudes and small viscosities. His result is given by

$$\tau_{\ell} = \frac{1}{(\ell - 1)(2\ell + 1)} \frac{R^2 \varrho}{\eta}$$
(4)

(where ρ is the density of the drop and η its dynamic viscosity).

Throughout the last 50 years, scientists studied how features such as viscosity, rotational flow and finite amplitudes influence the oscillation dynamics (as well as compressibility, which will not be discussed here). Miller and Scriven [32] considered the case of two incompressible, elastic and viscous fluids oscillating with small amplitudes. They derived transcendental equations governing the frequencies and damping rates. Later, Basaran *et al* [23] slightly corrected these calculations and found a damping rate of

$$\frac{1}{\tau} = \frac{(2\ell+1)^2 (\omega_\ell \eta_i \eta_o \varrho_i \varrho_o)^{1/2}}{2\sqrt{2}R(\varrho_i(\ell+1)+\varrho_o\ell)[(\eta_i \varrho_i)^{1/2}+(\eta_o \varrho_o)^{1/2}]} - \frac{(2\ell+1)^4 \eta_i \eta_o \varrho_i \varrho_o}{4R^2 (\varrho_i(\ell+1)+\varrho_o\ell)^2 [(\eta_i \varrho_i)^{1/2}+(\eta_o \varrho_o)^{1/2}]^2} + \frac{(2\ell+1)\{2(\ell^2-1)\eta_i^2 \varrho_i+2\ell(\ell+2)\eta_o^2 \varrho_o+\eta_i \eta_o [\varrho_i(\ell+2)-\varrho_o(\ell-1)]\}}{2R^2 (\varrho_i(\ell+1)+\varrho_o\ell)[(\eta_i \varrho_i)^{1/2}+(\eta_o \varrho_o)^{1/2}]^2}$$
(5)

(with the natural frequency ω_{ℓ} found by Lamb, equation (2)) in the limit of small viscosities. Prosperetti [33] derived equations governing the influence of viscosity on axisymmetric deformations of small amplitudes allowing vortex flow.

Tsamopoulos and Brown [22] were the first to investigate axisymmetric deformations of moderate amplitudes. They considered the special cases of drops $(\varrho_i \gg \varrho_o)$ and bubbles $(\varrho_i \ll \varrho_o)$ made of incompressible and non-viscous fluids. Expanding the surface, velocity potential and frequency in a Poincaré–Lindstedt manner in a small quantity ε , for example, the dimensionless surface

$$r(\theta, t; \varepsilon) = F(\theta, t; \varepsilon) = \sum_{k=0}^{\infty} \frac{\varepsilon}{k!} F^{(k)}(\theta, t) = 1 + \sum_{\ell=2}^{\infty} \varepsilon_{\ell} P_{\ell}(\cos \theta) \cos(\omega_{\ell} t) + O(\varepsilon^{2})$$
(6)

(P_{ℓ} are the Legendre polynomials in ℓ , which are proportional to the spherical harmonics Y_{ℓ} by a factor specified by ℓ), they derived the following dependence of the frequency on the amplitude, which is correct up to the fourth order in ε :

$$\omega_{\ell} = \omega_{\ell}^{(0)} \left(1 - \frac{1}{2} \omega_{\ell}^{(2)} \varepsilon^2 \right) \tag{7}$$

 $(\omega_{\ell}^{(0)}$ is the natural frequency given by equation (2)); the $\omega_{\ell}^{(2)}$ are given in table 1. Tsamopoulos [34] corrected these coefficients in 1989 (values in parentheses).

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In addition the authors found that an oscillation of the second mode in ε excites an oscillation of the zeroth mode and of the fourth mode with an amplitude of the order of $O(\varepsilon^2)$ and twice the frequency of the second mode [22]. The same happens when the fourth mode is oscillating in $O(\varepsilon)$. The odd numbered modes can excite even and odd modes. Furthermore, they showed that drops remain for a longer time in prolate than in oblate shapes (for a bubble both cases are almost equally distributed). Lundgren and Mansour [35] applied the boundary integral method to the problem of a drop undergoing axially symmetric oscillations accounting for weak viscous effects and found that an initial excitation of one single mode triggers oscillations of different modes with the frequency and twice the frequency of the exciting mode. In order to be able eventually to include viscous effects of arbitrary magnitude, Patzek *et al* [36] solved the Navier–Stokes equations of an incompressible and inviscid drop using a domain differential method. Lu and Apfel [25] calculated how the presence of surfactants changes primarily the damping of the small amplitude oscillations of an incompressible fluid immersed in another fluid using linear stability theory. Basaran [24] found numerically that damping is stronger for large amplitudes in the beginning of an oscillation.

Trinh *et al* [37] studied experimentally the axisymmetric oscillations of drops (radius $\approx 1 \text{ mm}$) made of silicon oil mixed with carbon tetrachloride in distilled water and drops made of phenetole in distilled water. The drops were held in the surrounding liquid by an acoustic field of high frequency, which was then modulated with a low frequency to drive the drop into the different modes of oscillation. The extracted eigenfrequencies of the different modes showed good agreement with Lamb's predictions (equation (2)). The damping rates were compared to an equation derived by Marston [38], which is equal to equation (5). Trinh *et al* [37] also found a dependence of the eigenfrequencies on the amplitude of deformation.

Becker *et al* [26] examined axisymmetric, free oscillations of drops (radius $\approx 0.2 \text{ mm}$), which periodically broke off a jet of ethanol (in air). After expanding the surface into a series of Legendre polynomials in $\cos(\theta)$, they found the eigenfrequencies and damping rates by fitting the amplitude versus time dependence with a function $\propto A_{\ell} e^{-t/\tau_{\ell}} \sin(\omega_{\ell}(t) t + \phi) + \beta$. In this way they accounted for the frequency change with time. Both the damping rates and the eigenfrequencies in the limit of very small amplitude agreed with Lamb; the frequencies increased with time (decaying oscillation amplitude).

Wang *et al* [39] regarded free axisymmetric oscillations of a drop (radius 1 cm) made of a glycerin/water mixture under microgravity conditions. The eigenfrequencies were in agreement with the values derived by Lamb. The frequency shift with the amplitude ε of the deformations agreed with the predictions of Tsamopoulos and Brown [22].

Most of the published material refers to theoretical descriptions of droplet or bubble oscillations. The experiment described in the following has been designed to test for the validity of various theoretical predictions.

3. Experimental setup and sample preparation

In figure 1(b), the experimental setup is sketched schematically. The soap bubbles are created in a container of acrylic glass, its bottom has been filled with butane gas. The density gradient allows the soap bubbles to float in the box without sinking to the bottom, for a time long enough so that collisions can be monitored. Above the container, a mirror was installed at an angle of 45° to the viewing direction. The camera observes both the direct front view of the soap bubbles and the mirrored top view in order to follow the bubble oscillations from two aspects.

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Figure 2. (a) Initial state of the soap bubble 08 shortly after the break of the contact film area, (b) initial amplitudes of the different deformation modes, equation (1), and (c) soap bubble 08 at maximum local deformation, the marked distance is equal to 1.2 cm.

The bubbles in the container and their mirror images are recorded with a high-speed camera (Photron Fastcam-ultima APX) at 1000–2000 fps (frames per second). In order to clearly detect the edge of the bubbles in each frame, the back and the bottom of the box were lined with black sheets, which reduced reflections from behind, and the soap bubbles were illuminated with four single cold light sources (Schott KL2500 LCD) from each corner of the box. In addition, the side walls were lined either with a white sheet or with creased aluminium foil to ensure good reflections onto the bubbles.

As the material for the bubble films we used a commercially available soap solution (Dr Rolf Hein GmbH & Co. KG) with a surface tension of $\sigma = 0.034$ N m⁻¹. The soap bubbles consist of thin films (the thickness is estimated from reflective colors to be less than $1 \mu m$; it varies over the surface); they are inflated with air. A density of air of $\rho_{i,o} = 1.2 \text{ kg m}^{-3}$ and a viscosity of $17.1 \times 10^{-6} \text{ kg m}^{-1} \text{ s}^{-1}$ are assumed. The mass of the inner gas ranges from 96 to 4.1 μ g, whereas the mass of the soap film (with film thickness of approximately 1 μ m) ranges from 8.6 to 1.0 μ g, respectively, for the largest and smallest bubbles. The Bond number is of the order of 10^{-3} ; therefore the equilibrium state of the bubbles is, in very good approximation, spherical. Since the frames taken with the camera represent two-dimensional projections of the three-dimensional motion, only bubbles with axisymmetric oscillations were selected for data analysis. In order to guarantee this we solely considered those cases in which two soap bubbles, initially spherical, approached each other and formed a common interface, which then tore apart (figure 1(c)). The fused bubble is very far from its equilibrium shape and the deformation is axisymmetric (see figure 2(a)). Therefore the bubble can be represented by a two-dimensional plot, which is then rotated around the symmetry axis to create the three-dimensional object. If the axis is normal to the observation direction, the entire information of the motion is contained in the front views. The top views were used to select those collisions only where the rotational symmetry axis was normal to the viewing direction. We have recorded more than 50 videos

Table 2. Experimental soap bubbles selected for this presentation, their equilibrium radii (radius of an equivalent sphere of the same volume) and frame rates in the videos.

Experiment	08	16	18	33
<i>R</i> (mm)	26.2	25.2	9.1	12.0
Frame rate (fps)	2000	2000	2000	1000

Table 3. Real-time duration and image width of the supplementary video material, available at stacks.iop.org/NJP/12/073031/mmedia.

Experiment = filename	08	16	18	33
Duration (s)	0.350	0.346	0.400	0.282
Width (mm)	74.7	70.1	34.7	71.4



Figure 3. Initial states of the soap bubbles 16, 18 and 33 (left to right) shortly after the break of the contact film area. Image widths are 54 mm (left), 35 mm (middle) and 36 mm (right).

of individual soap bubble collisions. For the present analysis in this paper, a few of them are showcased. Their motions included all important features.

4. Experimental results

In the following, the features of oscillating soap bubbles will be demonstrated with four sample experiments. They differ in their equilibrium radii as shown in table 2 and symmetries of the initial deformations (cf figures 2(a) and 3). The videos of the four soap bubbles selected for this presentation can be found in the supplementary material (see table 3).

In figure 2(a) one can see the state of the soap bubble 08 immediately after the fusion. From each image recorded during the subsequent oscillations, the contour of the bubble is detected with the help of a MATLAB program and $r(\theta)$ (figure 1(a)) is expanded in a series of spherical harmonics. The origin of the coordinate system is chosen at the center of mass of the inner



Figure 4. Area of soap bubble 08 as a function of time.

gas volume. In addition, the inner volume and the surface area are calculated. We found in our various experiments that the volume is conserved throughout the whole relaxation process, so the incompressible media models as described in section 2 should be applicable. Taking a look at figure 2(c), this is obvious: the image shows the instant of maximum local deformation during the motion. The local pressure caused by the film curvature following Laplace's law, $p = 2\sigma (R_1^{-1} + R_2^{-1})$ (with the radii R_1 and R_2 of the principal curvatures), is only of the order of about 20 Pa, almost four orders of magnitude smaller than the ambient pressure.

The expansion shows that the bubble geometry is dominated by the Y_0 mode, but that the amplitudes of the other modes are not small. In figure 2(b), the initial amplitudes of the first 13 modes are displayed for bubble 08. Since we are not interested in the fusion process itself, the exact moment of fusion is irrelevant. We refer to the initial amplitudes as the ones determined in the first image recorded immediately (i.e. within less than 1 ms) after the fusion. The second mode plays the most important role in the deformation. With higher mode numbers, the initial amplitudes decrease rapidly. The second mode is dominant because of the particular way the initial shape of the bubble was achieved: two spheres of rather comparable size merging. When two bubbles of equal size coalesce (cf bubble 33 in figure 3, right panel), there is even an additional symmetry element, a mirror plane perpendicular to the rotational symmetry with respect to this plane, namely the even numbered modes.

From this state very far from equilibrium, the soap bubble starts to oscillate. The oscillations are driven by the energy released during the transformation of two soap bubbles (surfaces 4000 and 6500 mm²) to one single soap bubble (equilibrium surface 8600 mm²). During the coalescence, the surface area performs rapid oscillations (see figure 4). The motions of the zeroth, second, third and fourth modes are displayed in figure 5. Figure 5(b) shows how the second mode seems to be oscillating in a simple cosine-like manner. A closer look reveals that the amplitude remains positive for a longer portion of the time, namely 58.7%. Hence the



Figure 5. Amplitudes of the different modes (expansion coefficients of the spherical harmonics) of soap bubble 08 as a function of time.

bubble remains for a longer time in the prolate shape than in the oblate one. This was already shown by Tsamopolous and Brown [22] and various other authors for liquid droplets and gas bubbles in a liquid. The evolution of the zeroth mode in figure 5(a) is quite different: at first there is a fast process where the bubble is moving closer to its equilibrium state. After 5 ms, an oscillation is observable with multiple frequencies. This originates from the condition of volume conservation, equation (3). When the amplitudes of the higher modes start to oscillate, the volume would change unless the zeroth mode starts to oscillate synchronously with the frequencies of the other modes. This can be illustrated, e.g., by a calculation of the bubble volume when only the second mode is excited, with amplitude A

$$V = \frac{4\pi}{3}R(t)^3 + R(t)A(t)^2 + \frac{\sqrt{5}}{21\sqrt{\pi}}A(t)^3 = \text{const.}$$
(8)

R(t) is not equal to the equilibrium radius; it approaches it only when the amplitude A of the second mode decays to zero. At preserved bubble volume, R is independent of A(t) only in linear approximation. Hence, the radius R (zeroth mode) has to change in time with the amplitude of the deformation. Since the second and third modes are the dominant ones in the oscillation, their natural frequencies should be present in the dynamics of the zeroth mode. This

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Figure 6. Windowed Fourier transformation of the second mode, figure 5(b), and of the fourth mode, figure 5(d), soap bubble 08.

is different from the linearized approximation (in equation (8) the amplitudes of deformation are present in higher order). A similar result was found theoretically by Basaran [24], Mashayek and Ashgriz [40] and Shaw [31]. Their descriptions are basically in good agreement with our observations, although a direct comparison is difficult. The dynamics of the fourth mode reveals that the oscillation is modulated with multiple frequencies (figure 5(d)). In addition, the amplitude is positive for a great portion (83.7%) of the time. In order to analyze the evolution of the frequency spectrum, a windowed fast Fourier transformation (FFT) was performed with each dataset (for the second and fourth modes, this is shown in figures 6(a) and (b)). The width of the Hamming window was 160 ms. This window was shifted in steps of 5 ms in order to obtain time-resolved spectra. The amplitude of the Fourier transform was scaled by the number of points in each FFT. Figure 6(a) shows that one frequency is dominant in the second mode; it is just damped with time. The damping constant (see table 8) can be extracted from the time decay of the integral of a given Fourier peak. In figure 7 the amplitude change of the eigenfrequency belonging to the second mode of bubble 18 is displayed. It reveals that damping is slightly higher in the beginning, which was found theoretically by Prosperetti [33] and Basaran [24]. An exponential fit with a mean damping rate is displayed by a solid (red) line. The damping constants will be discussed in more detail in the next section.

A closer look at the Fourier transforms reveals that the peak frequency of the second mode (the eigenfrequency) changes slowly with time, which is displayed in figure 8(a). It increases with time, which indicates that it increases with decreasing amplitude. As Tsamopoulos and Brown [22] derived equation (7), we define an equivalent ε' here, namely the amplitude of the peak frequency as found in the Fourier transform in figure 6(a) divided by the equilibrium bubble radius. The quantity ε' differs from the ε (see equation (6)) of Tsamopoulos and Brown only by a constant factor, since here the amplitudes of spherical harmonics are considered as opposed to their Legendre polynomials. For the second mode, $\varepsilon' = 2\sqrt{\pi/5} \varepsilon$, and for the third mode, $\varepsilon' = 2\sqrt{\pi/7} \varepsilon$. The change of frequency with ε' is plotted in figure 8(b), which shows a qualitatively similar behavior as that predicted in [22]. A quadratic fit of the form $f = f_0(1 - \frac{1}{2}\tilde{\omega}^{(2)}\varepsilon'^2)$ enables us to find the coefficients $\omega_{\ell}^{(2)}$. It is

A quadratic fit of the form $f = f_0(1 - \frac{1}{2}\tilde{\omega}^{(2)}\varepsilon'^2)$ enables us to find the coefficients $\omega_{\ell}^{(2)}$. It is indicated by the solid line in figure 8(b). A clearer presentation of the qualitative behavior of the



Figure 7. Change of the Fourier amplitude with time for the eigenfrequency of the second mode (maximum amplitude in the FFT), soap bubble 18.



Figure 8. Change of the frequency of the second mode with time and amplitude, respectively (the arrow indicates the direction of time), soap bubble 08.

frequency is displayed in figure 9 for soap bubble 18, which was recorded until the oscillations had essentially died out. The graph shows the asymptotic frequency limit that is reached for very small amplitudes. Note that the values for the smallest ε' correspond to an oscillation amplitude of only 2% of the bubble radius; the determined frequencies are affected by larger experimental uncertainties than the remaining curve. The results for all bubbles are summarized in table 4.

When these values are compared to those in table 1, one notices considerable deviations. A possible reason for the difference could be that in the model of Tsamopoulos and Brown, $\omega_{\ell}^{(2)}$ has been derived for only one excited mode with amplitude ε . In our case there are several oscillation modes from the beginning, so an energy transfer between the modes is possible and the value of ε' is affected. In addition, ε is taken as a moderate quantity in the theoretical





Figure 9. Change of the frequency of the second mode with amplitude ε' , soap bubble 18.

Table 4. Values of the quadratic coefficients $\omega_{\ell}^{(2)}$ of the frequency shift for each soap bubble. Bubbles 18 and 33 were formed by two almost equal fusing bubbles; thus they are almost mirror symmetric with respect to a plane perpendicular to the central axis. The odd modes are hardly excited then.

	08	16	18	33
$\ell = 2$	2.3	2.0	1.6	1.5
$\ell = 3$	6.0	7.1	-	-

model, while the oscillations in our experiments are of large amplitude. This complicates a comparison.

From a construction of the eigenfrequency versus ε' for each mode (whenever possible) and each soap bubble, we find the limit of the eigenfrequencies for $\varepsilon' \to 0$, i.e. for small amplitudes. The results are summarized in table 5. Normal numbers are the extrapolated frequencies for $\varepsilon' = 0$ in the fit $\omega_{\ell}/(2\pi) = f_{\ell} = f_{\ell}^{(0)}(1 - \frac{1}{2}\tilde{\omega}_{\ell}^{(2)}\varepsilon'^2)$ and italic numbers are frequencies estimated from the trend of the graphs. Since the higher modes are excited with very small amplitudes, a quadratic fit is not useful there. The linear eigenfrequencies calculated with the adapted equation (2)

$$\omega_{\ell} = \sqrt{\frac{2\sigma}{R^{3}\varrho} \frac{(\ell-1)\ell(\ell+1)(\ell+2)}{2\ell+1}}$$
(9)

(two film surfaces, equal densities $\rho_i = \rho_o = \rho$ of the inner and outer media) are included for comparison. It has to be noted that in the spectra of the higher modes (cf figure 6(b)), multiple peaks are found. In these spectra, one has to neglect frequencies that appear as a consequence of nonlinear coupling, in order to select the eigenfrequency. The nonlinear coupling effects are discussed below.

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uncer	incertainty of ±1.5 Hz for each frequency.									
	08		16		18		33			
	Experiment	Lamb	Experiment	Lamb	Experiment	Lamb	Experiment	Lamb		
$\ell = 2$	19	19.57	19	20.75	85	95.45	70	63.14		
$\ell = 3$	34	36.99	35	39.21						
$\ell = 4$	51	56.50	57	59.90	240	275.53	202	182.28		
$\ell = 5$	71	78.07	72	82.76						

Table 5. Asymptotic frequencies $f_{\ell} = \omega_{\ell}/(2\pi)$ and theoretical values of Lamb [21] for each soap bubble. All frequencies are given in Hz. We estimate an uncertainty of ± 1.5 Hz for each frequency.

Table 6. Ratios of different eigenfrequencies. The uncertainty of the experimental ratios is $\pm 10\%$.

Eigenfrequencies	Ratio according to equation (9)	08	16	18	33
Mode 3/mode 2	1.89	1.81	1.83	_	_
Mode 4/mode 2	2.89	2.70	2.96	2.87	2.82
Mode 5/mode 2	3.99	3.75	3.79	_	_
Mode 4/mode 3	1.53	1.49	1.62	_	_
Mode 5/mode 3	2.11	2.07	2.07	_	_
Mode 5/mode 4	1.38	1.39	1.28	-	-

The experimentally extrapolated eigenfrequencies agree satisfactorily with those derived from Lamb's linear model. Deviations from the model may have several reasons. For example, the experimental uncertainty of the bubble volume (equilibrium radius), experimental uncertainties in the determination of the surface tension or others may affect the absolute frequencies. In order to eliminate these effects, we compare the ratios of the different eigenfrequencies in table 6 with those predicted by Lamb. This is useful because, according to formula (2), the parameters radius, surface tension and density cancel. These data are reported in table 6.

All ratios coincide within experimental uncertainty of a few per cent. In conclusion, Lamb's formula is correct for soap bubble oscillations at small amplitudes, but the frequencies decrease quadratically with increasing oscillation amplitudes. The deviations of the absolute values will be discussed in the last section.

After focusing on the eigenfrequencies, we discuss now the complete spectra (figure 5). The Fourier transform of the fourth mode in figure 6(b) has an appearance qualitatively very different from that of the second mode. There are at least three different frequency peaks present: one near the eigenfrequency of the second mode (≈ 18 Hz), one near the eigenfrequency of the second mode (≈ 18 Hz), one near the eigenfrequency of the fourth mode is the one with the fourth mode (≈ 51 Hz). Initially, the natural frequency of the fourth mode is the one with the highest amplitude but with time the natural frequency of the fourth mode is not simply damped exponentially, but has a peak at about the same time as the third mode. This evidences very

Table 7. Time-averaged dominant frequencies $\omega/2\pi$ (Hz) in the spectra of individual modes; bold numbers are the natural frequencies of each mode; each frequency is afflicted with an uncertainty of ± 1.5 Hz; the numbers in parentheses represent the largest amplitude (mm s) of each frequency peak in the Fourier transforms.

	08	16	18	33
$\ell = 0$	18 (0.17)	19 (0.08)	167 (0.08)	122 (0.03)
	36 (0.24)	36 (0.14)		
	50 (0.03)			
$\ell = 1$	16 (0.23)	16 (0.13)	83 (0.01)	-
	33 (0.11)	35 (0.05)		
	51 (0.22)	53 (0.14)		
$\ell = 2$	18 (3.11)	19 (2.6)	83 (1.2)	70 (0.8)
0 2	15 (0.22)	15 (0.22)		
$\ell = 3$	15 (0.22)	15 (0.22)	_	_
	33 (1.2)	35 (0.81)		
	50 (0.16)			
	69 (0.21)			
$\ell = 4$	18 (0.14)	18 (0.08)	156 (0.1)	140 (0.03)
	35 (0.17)	37 (0.11)	239 (0.12)	200 (0.05)
	51 (0.17)	55 (0.07)		
$\ell = 5$	15 (0.15)	73 (0.25)	_	_
	33 (0.1)			
	70 (0.35)			
	88 (0.15)			

complex behavior of the mode coupling, which is of course not contained in the linear model. Table 7 includes the most important frequencies that are present in the spectra of the individual modes (more frequencies may be present with lower amplitudes). They have been obtained by means of Fourier transformation of the complete observation period and thus represent some average value for each mode, neglecting the $\approx 5\%$ shift with varying amplitude. Therefore, they may not be compared directly with the asymptotic frequencies in table 5 that represent the small amplitude limits.

Table 7 shows the very complex coupling behavior between the modes, as already discussed earlier. Our assumption about the motion of the zeroth mode (figure 5(a)) is confirmed: the frequencies of the most important modes are found in its spectrum, confirming that the oscillation of the zeroth mode is required for the conservation of volume. An interesting effect can be seen in soap bubbles 18 and 33. Their initial deformation was almost symmetric so that initially only the even numbered modes were excited. Thus the odd modes remained essentially zero all the time. It is obvious that for symmetry reasons, oscillations that contain only even modes can excite only even modes, whereas odd modes, in connection with the zeroth mode, can couple to both odd and even modes.

All spectra of the fourth mode contain peaks at twice the natural frequency f_2 of the second mode. In bubbles 08 and 16, it is difficult to discriminate this from a coupling to the third mode, which has an eigenfrequency f_3 rather close to $2f_2$. However, for bubbles 18 and 33, it can be

clearly attributed to a coupling between the second and fourth modes, since the third mode is practically absent. Such coupling was first reported for droplet oscillations by Tsamopoulos and Brown [22] and later by Lundgren and Mansour [35]. The oscillation of the second mode acts on the fourth mode as an excitation of second order. It results in a frequency peak of twice the driving frequency.

It is important to note that in bubbles 08 and 16, the natural frequency f_2 of the second mode appears in the odd numbered modes reduced by about 2 Hz. In return, the natural frequency f_3 of the third mode appears in even numbered modes increased by about 2 Hz. This shift reflects a resonant excitation in the different cases, since f_3 is almost twice f_2 but not quite (table 6). This effect was already reported experimentally for liquid bubbles by Trinh *et al* [41]. It is, however, difficult to distinguish from the quadratic coupling effect mentioned above. Since the frequency ratios of the third to fifth modes to that of the second mode are very close to integers (2, 3 and 4), coupling is very efficient.

As one can see in table 7, one also finds a weakly excited but non-zero first mode. This is due to the fact that the center of mass of the inner gas is set to be the origin of the coordinate system. Owing to this choice, the first mode oscillates to compensate for a displacement of the centroid by the large-amplitude oscillations of higher modes. The oscillations of the center of mass in the laboratory fixed frame (not shown) are correlated to the oscillations of every mode (as Shaw [31] showed for gas bubbles in an ambient liquid).

5. Simulation

In order to compare the experimental results with the exact solution of the Navier–Stokes and continuity equations, a numerical simulation of the soap bubbles was performed. Details of this calculation will be published elsewhere. A mapped finite elements method on interface-resolving moving meshes was used to solve the incompressible Navier–Stokes equation [42]

$$\rho \left[\frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \nabla) \vec{v} \right] = -\text{grad } p + \eta \, \triangle \vec{v}, \tag{10}$$

$$\operatorname{div} \vec{v} = 0. \tag{11}$$

The considered problem has axial symmetry. Therefore a three-dimensional axisymmetric formulation has been used in our computations [43]. Note that the accuracy and performance of a two-dimensional plane variant of the code have been evaluated in quantitative benchmark computations [44]. Calculations were performed in dimensionless coordinates. The proper scaling was realized through the Reynolds, Froude and Weber numbers, taking into consideration the surface tension of the film, density and viscosity of the air and bubble dimensions. In the simulation we consider two incompressible media, one surrounding the other. Both are separated by a freely movable interface with a surface tension but otherwise negligible physical parameters (inertia, viscosity and elasticity). The dimensions of the simulation box were chosen large enough so that there was no relevant flow near the outer boundaries. The boundary condition at the interface (bubble surface) is chosen such that the flow field is continuous across the liquid film. This means, in particular, that the flow normal to the film is the same on both sides. Flow in the film plane advects the liquid film material in the experiment. However, there is no flow in the film plane normal to the plane formed by the symmetry axis



Figure 10. Change of the amplitude with time of the second mode for (a) experiment 08 and (b) the simulation of soap bubble 08.



Figure 11. Change of the amplitude with time of the fifth mode for (a) experiment 08 and (b) the simulation of soap bubble 08. We show only a short time interval, to resolve more details.

and the radial coordinate (no tangential flow) in the simulation. The initial shape of the interface can be chosen arbitrarily in the program. The motion starts from a deformed bubble at rest.

In order to simulate the experimental observations, we have used the parameters for air and twice the surface tension of the employed soap solution (two film surfaces). We have imported the rescaled interface data of the experimental soap bubble 08, figure 2(a), as the initial conditions. The calculated shapes of the interface have been evaluated to extract amplitudes of the oscillation modes (spherical harmonics) as a function of time. This allows for direct comparison with the experiment. Two sample graphs (the second and fifth modes of bubble 08) are shown in figures 10 and 11.

The Fourier transforms of the simulated data reveal that the frequencies obtained from the simulation agree with the experiment within the resolution of the measurement. Figure 11 shows that even details of the complex time-dependent amplitudes are reproduced, except for the

	08		16			18		33		
	Exp.	Sim.	Basaran <i>et al</i>	Exp.	Basaran <i>et al</i>	Exp.	Basaran <i>et al</i>	Exp.	Basaran <i>et al</i>	
			[23]		[23]		[23]		[23]	
$\ell = 2$	0.36	0.57	0.70	0.38	0.65	0.081	0.109	0.095	0.177	
$\ell = 3$	0.19	0.33	0.36	0.22	0.34	_	-	_	-	
$\ell = 4$	-	-	_	-	_	0.046	0.036	0.030	0.058	

Table 8. Experimental and theoretical (equation (5)) damping rates (seconds) for every soap bubble and damping rates from the simulation of bubble 08.

damping rates. In both figures 10 and 11, the oscillations relax much more quickly in the experiment. The damping rates are summarized in table 8. In addition, the theoretical values given by Basaran *et al* [23] are included in that table. The damping constants obtained by the simulation are slightly smaller than the theoretical values. A comparison of the experimental damping rates with those derived by Basaran shows that for all soap bubbles and almost every mode the experiment damps twice as quickly as predicted by theory. The only exception is the fourth mode of soap bubble 18, but in this case we have to assume that this mode is permanently excited by nonlinear coupling. Miller and Scriven [32] have considered the influences of the membrane by assuming inextensibility of a liquid film separating two fluids. They found a higher damping than Basaran *et al* (but still lower than our experimental results). In our soap films the condition of inextensibility does not hold since figure 4 shows rapid surface oscillations.

6. Conclusions and summary

The experiments were designed to test the validity of the analytical linearized model (Lamb) that makes several simplifying assumptions and subsequent theoretical work that has included nonlinear effects in the dynamic equations. Before we discuss the detailed results, a few general remarks are necessary. The first concerns a purely technical point; the bubbles have been observed for time periods between 0.3 and 0.4 s. This implies that the frequencies reported here can only be resolved with an accuracy of 1.67 and 1.25 Hz, respectively. A better resolution cannot be achieved. Even if the bubbles are observed for longer times, the oscillation amplitudes will have decayed below detection level. Therefore, there is a limit in principle of the resolution of the eigenfrequencies, which can only be circumvented when the damping rates are reduced. Even the replacement of air as the outer and inner media by a gas of lower viscosity (H_2) would not help, since the ratio of density and viscosity is relevant.

Secondly, we need to discuss an experimental problem, the presence of butane in the environment of the bubbles. We use this gas to keep the bubbles in the viewing field of the camera during the fusion experiment. If the outer medium were pure butane and the inner medium were pure air, the experimental results would be essentially different from our observations. However, the bubble floats on a mixture of air with a low butane content. A back-of-the-envelope calculation shows that the liquid film has a mass that is a few per cent of the bubble mass (e.g. less than 10% for bubble 08). The buoyancy in the outer gas approximately compensates for the weight of the liquid film; thus the outer medium will have a density that is approximately 10% higher than the inner gas. One can see immediately, e.g. from equation (2), that the effect on the eigenfrequencies is at the limits of our experimental resolution. The relative

influence on the frequencies in equation (2) reduces to 2.5%. It may account for the fact that we measure systematically too low frequencies (table 5). For bubble 18, the smallest of all, the liquid film may contribute up to 25% of the bubble weight. This means that we underestimate the density of the outer liquid in the calculations by approximately 25%, and the calculated frequency is overestimated by about 6%. As one can see in table 5, the deviations of measured frequencies from Lamb's model are largest for this bubble. Another reason for overestimated frequencies is the neglecting of the film mass in the analytical model. Its influence may be of the same order of magnitude as the previous correction.

The film thickness has been estimated from bubbles observed in parallel, reflected light. We found that the thickness of the bubbles produced in our setup was statistically distributed in the submicrometer range, from a few hundred nanometers to about $1 \mu m$. The properties of the liquid membrane (except the surface tension) can be neglected for bubbles much larger than 1 cm in diameter when one is interested in the frequency spectrum, but the membrane seems to be responsible, among other effects, for a slightly (10%) lowered oscillation frequency of the smallest bubble reported in this study. The most important influence of the liquid film obviously manifests itself in the comparably large experimental damping rates. Damping of the bubble oscillations is stronger than both the prediction by the numerical simulation and the analytical values (cf table 8). This indicates that the viscosity of the liquid film, which is not considered in the computations, contributes significantly to the dissipation of energy. Thus, the film motion seems to contribute significantly to the dissipation processes. Its quantitative evaluation is difficult, particularly because the films are never uniformly thick, but it seems possible to study this influence by manipulating the film viscosity, e.g. by the addition of glycerol.

Summarizing, the experiments have shown that the linearized analytical treatment by Lamb describes the observed eigenfrequencies of the oscillation modes correctly. In addition, the nonlinear corrections have at least three effects that are not predicted by the linear model. We find a decrease of the eigenfrequencies as a consequence of nonlinear terms at large amplitude oscillations. The shift of the frequencies of the modes with increasing amplitude agrees with the quadratic prediction by Tsamopoulos and Brown [22], equation (7). However, the coefficients are found to be quantitatively larger than predicted (table 4). Another nonlinear effect described in the literature for droplets and bubbles is the coupling of individual modes. This effect has been confirmed in our experiments. In particular, the frequency of the second mode is found in almost all other modes. Nevertheless, there are two limitations to the coupling between the eigenmodes. Firstly if we start with a symmetric deformation (fusion of two equally sized bubbles), only even numbered (symmetric) modes are excited. Secondly, since the initial state is axially symmetric, the bubble oscillations retain axial symmetry until equilibrium is reached. During the rapid decay of the initial deformations, non-axisymmetric modes (spherical harmonics Y_l^m with non-zero second index *m*) are not measurably excited.

The third consequence of nonlinearity is the observed oscillation of the zeroth and first modes. The latter result is peculiar for our systems with nearly equal densities of the inner and outer media.

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