

Nonlinear free-decay oscillations of a magnetically levitated air bubble in water produced by coalescence

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The moderate- to large-amplitude free-decay oscillations of an air bubble in water, formed from the coalescence of two parent bubbles, are studied experimentally and by numerical simulation. Magnetic levitation is used to suspend the air bubble, with a diameter of approximately 6 mm, within the liquid as if under weightless conditions. We find that the method does not significantly distort the shape of the air bubble, facilitating direct comparison with theoretical predictions of oscillations from an assumed spherical equilibrium shape. The coalescence of two equal-sized parent bubbles produces a single bubble undergoing freely decaying nonlinear shape oscillations dominated by the two-lobed shape mode, approximating the problem considered theoretically by Tsamopoulos and Brown, who obtained the corrections to the shapes and frequencies of the linear modes of oscillation to second and third order in amplitude, respectively. The frequency of the two-lobed oscillations decreases with the square of the amplitude, as predicted, but with a coefficient smaller in magnitude than expected. Coalescence of two unequal-sized bubbles, differing in diameter by a factor 1.54, produced a bubble oscillating with comparable contributions from two- and three-lobed shapes. Here the quadratic coefficient is the same (within uncertainty) as that observed in the symmetric case. However, there is also a significant dependence of the frequency on the fourth power of the amplitude in this case, and the quadratic coefficient of the three-lobed mode is approximately three times larger than predicted, which we attribute to coupling between the two- and three-lobed modes. The magnitudes of the various surface harmonics contributing to the shape of the two-lobed mode agree well with Tsamopoulos and Brown's prediction at second order in amplitude and with our calculations at third order. Results from simulation are consistent with those from experiment.

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I. INTRODUCTION

The nonlinear axisymmetric shape oscillations of a bubble of gas in liquid have been studied theoretically by Tsamopoulos and Brown (T&B) [1], who considered the moderate-amplitude two-, three-, and four-lobed oscillations [2] corresponding to the three lowest order linear shape oscillation modes $n = 2-4$ [3]. Moderate- to large-amplitude bubble-shape oscillations frequently arise from

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the coalescence of two smaller bubbles, in both natural and industrial processes. The resulting oscillations typically comprise several such nonlinear modes of oscillation with amplitudes that are a significant fraction of the bubble radius, and so effects of interaction between them might be expected. Here we study the nonlinear oscillations of a newly coalesced air bubble experimentally, using magnetic levitation to suspend the bubble freely in liquid by counteracting the buoyancy of the bubble, preventing it from rising.

The ability to acoustically “levitate” small bubbles of gas in liquid has been known for many years (e.g., [4]), using acoustic forces to balance their buoyancy. The same forces also provide a restoring force in all directions, trapping the bubble in place, thus permitting studies of isolated bubbles. One drawback of this method is that the equilibrium shape of bubbles trapped this way becomes increasingly nonspherical with increasing size [5]. Optical tweezers have also been used to study coated microbubbles, in which single or multiple bubbles are trapped by a tightly focused laser beam [6,7], but the size of bubbles that can be levitated by this method is limited to a few micrometers in diameter. These restrictions have motivated researchers to perform experiments in microgravity conditions in orbit (e.g., [8]). Here we present an alternative method of suspending air bubbles in water using magnetic levitation. We demonstrate that the technique can suspend quite large air bubbles, up to approximately half a centimeter in diameter, with insignificant distortion of the bubble shape. Since the frequency f of the shape oscillations scales with radius R as $f \sim R^{-3/2}$, the use of bubbles of this size allows us to obtain high-resolution images with a relatively high frame rate (more than 100 frames per period of oscillation of the lowest frequency mode), avoiding aliasing resulting from low sampling rate [9].

We consider two cases: a symmetric coalescence of two parent bubbles of equal size, and an asymmetric coalescence of parent bubbles differing in radius by a factor of 1.54. We obtain the frequency spectra of the spherical surface harmonics of the shape oscillations resulting from each of the two coalescences and compare these with the theory of T&B [1], who calculated the shape to second order in amplitude, and also with our calculations that extend this work to third order in amplitude. We compare theory and experiment with numerical simulations.

The deformation and breakup dynamics of gas bubbles plays a critical role in diverse natural and industrial processes. This is evident, for example, in large-scale phenomena such as the transport of gas released during oil well blowouts (e.g., [10,11]) and in CO₂ release in limnic eruptions (e.g., [12,13]), where these dynamics become more important with increasing bubble size (e.g., [14,15]). Bubbles are also employed in industrial processes to enhance mixing and mass transfer [16]. Knowledge of the oscillation modes of bubbles is fundamental for understanding their deformation dynamics. Historically, attention has focused on the volume- or “breathing”-mode oscillations [17], responsible for sound generation in liquids with entrained bubbles, producing, for example, the familiar noises made by a “babbling brook” or breaking waves. More recently, increased attention has been given to the shape oscillations of bubbles, where surface tension provides the restoring force. Longuet-Higgins showed that shape oscillations couple nonlinearly to volume oscillations, thus also contributing to sound generation [18] (see also [9,19–22]), and their stability is crucial in the phenomenon of sonoluminescence [23]. Shape oscillations are also relevant to the use of bubbles as contrast agents in medical ultrasound imaging and for targeted drug delivery (see, e.g., [24] and references therein). Returning to industrial applications, the shape oscillations significantly increase the rate of mass transfer in bubble column reactors [25,26].

In the above-mentioned examples, the amplitude of the shape oscillations is often a significant fraction of the radius of the bubble, such that the oscillations are nonlinear. Lamb, using linear stability analysis, calculated the frequencies of the normal modes of shape oscillations (i.e., of vanishingly small amplitude) for a spherical volume of inviscid fluid immersed in a second similar fluid [3]. Each mode, n , oscillates at a frequency dependent on the coefficient of surface tension, γ , the density of the inner and outer fluids, D_1 and D_2 , respectively, and the radius of the bubble at rest, R . If the density of the surrounding fluid is taken to be negligible in comparison to the density of the inner fluid, the earlier result derived by Lord Rayleigh [27] for the oscillation frequencies of a liquid drop is recovered, whereas if the density of the inner fluid is taken to be negligible in comparison to

the density of the surrounding fluid, the oscillation frequencies of a gas bubble in liquid are found,

$$\omega_n^{(0)} = \frac{1}{\tau} \sqrt{(n^2 - 1)(n + 2)}, \quad (1)$$

where $\tau = \sqrt{\frac{D_2 R^3}{\gamma}}$ is the capillary timescale. Here the superscript (0) denotes the leading order approximation to the oscillation frequency in the limit of vanishingly small perturbations to the bubble surface. T&B [1] used the Poincaré-Lindstedt method [28] to calculate corrections for the velocity potential, shape functions, and eigenfrequencies of individual axisymmetric modes of inviscid, incompressible bubbles, and drops. Expressing the shape of the bubble in polar coordinates $r(\theta)$, they obtained the corrections to the shapes of the linear modes of oscillation $n = 2-4$ to second order in amplitude:

$$\frac{r_n(\theta, t)}{R} = \sum_{k=0}^{\infty} \frac{\varepsilon^k}{k!} F_n^{(k)}(\theta, t) = 1 + \varepsilon F_n^{(1)}(\theta, t) + \frac{\varepsilon^2}{2} F_n^{(2)}(\theta, t) + O(\varepsilon^3), \quad (2)$$

where

$$F_n^{(1)}(\theta, t) = P_n(\theta) \cos(\omega_n t) \quad (3)$$

is the linear approximation to the shape function obtained by Lamb [3], and the second-order correction to the shape has the form

$$F_n^{(2)}(\theta, t) = \sum_{l \in S} c_{n,l} P_l(\theta) [\cos(2\omega_n t) + d_{n,l}]. \quad (4)$$

Here θ is the polar angle, ε is the dimensionless amplitude of oscillation, P_l is a Legendre polynomial of degree l , and ω_n is the amplitude-dependent frequency of the mode. Throughout we use the symbol n to refer to the mode of oscillation and the symbol l when referring to the degree of a surface harmonic. In (4), $c_{n,l}$ and $d_{n,l}$ are mode-dependent coefficients determined by T&B, and the sum runs over $S = \{0, 2, \dots, 2n\}$; we see from this equation that the corrections at second order contribute additional even-degree surface harmonics to the shape that oscillate at twice the frequency of the linear term. Extending the calculation of T&B to third order in ε introduces additional surface harmonics that oscillate at ω_n and $3\omega_n$ (Supplemental Material, Sec. SI [29]):

$$F_n^{(3)}(\theta, t) = \sum_{l \in S} e_{n,l} P_l(\theta) [(1 + f_{n,l}) \cos(\omega_n t) + f_{n,l} \cos(3\omega_n t)]. \quad (5)$$

Here the sum runs over $S = \{0, 2, 4, \dots, 3n\}$ for even n and $S = \{1, 3, 5, \dots, 3n\}$ for odd n , and $e_{n,l}$ and $f_{n,l}$ are mode-dependent coefficients calculated in the Supplemental Material (Sec. SI [29]). Expanding ω_n with ε ,

$$\omega_n = \sum_{k=0}^{\infty} \frac{\varepsilon^k}{k!} \omega_n^{(k)}, \quad (6)$$

the first- and third-order frequency coefficients, $\omega_n^{(1)}$ and $\omega_n^{(3)}$, were found to vanish [1],

$$\omega_n = \omega_n^{(0)} + \frac{\omega_n^{(2)}}{2} \varepsilon^2 + O(\varepsilon^4). \quad (7)$$

In the case of bubbles, the second-order frequency coefficient was determined by T&B to be $\omega_2^{(2)} = -1.526\omega_2^{(0)}$ and $\omega_3^{(2)} = -2.305\omega_3^{(0)}$ for $n = 2$ and 3 , respectively [30].

A shift to lower frequency of the two-lobed ($n = 2$) mode of liquid drops with increasing amplitude was observed by Wang *et al.* [31], who conducted experiments in microgravity, creating large initial perturbations using an acoustic trap, and by Becker *et al.* [32], who studied the large-amplitude oscillations of free-falling ethanol drops. Both observed the change in frequency

as a function of amplitude to be in good agreement with the predictions of T&B. Trinh *et al.* [33] observed the moderate-amplitude oscillations of acoustically trapped silicone oil drops in water. In experiments on drops driven into two-lobed shape oscillations, the presence of an $l = 4$ surface harmonic oscillating at a frequency approximately twice that of $\omega_2^{(0)}$ was observed, consistent with T&B's prediction.

Regarding experimental studies of the shape oscillations of bubbles, Asaki *et al.* [5] studied small-amplitude shape oscillations of relatively large bubbles, up to 12 mm diameter, using an acoustic levitator. Measurements of the frequency of two-lobed oscillations deviated from Lamb's theory, which the authors speculated was due to the oblate distortion of the bubble by the levitator; experiments performed in orbit appeared to confirm this [8]. Experiments studying the free decay of small-amplitude $n = 2$ oscillations have investigated the deviation of their frequency from Lamb's result (1) due to viscous damping [34]. Experiments on acoustically trapped air bubbles in water [33] studied oscillations of moderate amplitude, but the data do not show quantitative agreement with the results obtained by T&B. One reason for this may again be the oblate distortion of the bubble introduced by the acoustic levitator [35]. Other studies have focused on the coupling between shape and breathing modes [9,21,22,36].

Kornek *et al.* [37] studied the large-amplitude shape oscillations of thin-film soap bubbles (i.e., enclosing and surrounded by a gas) and found a quadratic relationship between the frequency and amplitude. In the case where two-lobed oscillations of large amplitude were excited, the second-order frequency coefficient, $\omega_2^{(2)}$ in (7), was found to be approximately 10% greater in magnitude than the value predicted by T&B, but in the case where both two- and three-lobed oscillations were excited with large amplitude, $\omega_2^{(2)}$ was found to be up to 60% greater, and the second-order coefficient of the three-lobed mode, $\omega_3^{(2)}$, was found to be more than three times T&B's value.

Shape oscillations excited by coalescence have been previously investigated in experiments on acoustically trapped bubbles with radii of the order 100 μm [9]. In those experiments, in which the oscillations were sustained by the acoustic field, the frequency of the breathing mode oscillations was comparable to those of low order ($n \leq 4$) shape modes, and coupling with the breathing mode oscillations was observed as a result (see also [22]). Here we study the free decay of shape oscillations resulting from the coalescence of bubbles with radii of a few millimeters (similar in size to those of bubbles found in a typical bubble column reactor), in which the breathing mode lies at a frequency $\omega_b/2\pi \sim 1$ kHz, well above those of the low-order shape modes, $\omega_n^{(0)}/2\pi \approx 30\text{--}100$ Hz for $n = 2\text{--}4$, in the bubbles studied here.

It has been known since 1939 that materials commonly thought of as “nonmagnetic” can be levitated magnetically, when Braunbek demonstrated the levitation of beads of graphite using an electromagnet [38,39]. The force lifting the graphite arises by virtue of graphite's diamagnetism, a weak form of magnetism which causes it to be repelled from a magnetic field with a force proportional to the strength of the magnetic field $B = |\vec{B}|$ and its gradient ∇B . Like gravity, this force acts as a long-range force throughout the body of the object rather than acting at its surface, in contrast to other methods of levitation. In the 1990s other diamagnetic materials including water, plastics, biological organisms and tissues, and liquid helium were demonstrated to levitate in strong magnetic fields generated by resistive or superconducting solenoids [40–43]. Diamagnetic levitation typically requires a strong magnetic field with a large spatial gradient. To levitate water, for example, requires $|B \partial B / \partial z| \approx 1500 \text{ T}^2 \text{ m}^{-1}$, where $\partial B / \partial z$ is the vertical gradient of B . Immersion in a paramagnetic fluid allows suspension of diamagnetic materials in weaker magnetic fields (“magneto-Archimedes levitation”) [44–47] and has also been used in developing magnetic noncontact self-assembly techniques (e.g., [48,49]).

The sum of the magnetic and gravitational potential energy (magnetogravitational potential) of an object immersed in a fluid at a point $\vec{x}$, in a magnetic field with spatially varying strength $B(\vec{x})$, is given to a good approximation by ([41,43,44], see also Sec. SII [29])

$$E_{\text{MG}}(\vec{x}) = \frac{(\chi_2 - \chi_1)}{2\mu_0} V B(\vec{x})^2 - (D_2 - D_1) V g z. \quad (8)$$

Here V is the volume of the object, z is the vertical coordinate, g is the acceleration due to gravity, μ_0 is the magnetic constant, and D_j and χ_j are the densities and volume magnetic susceptibilities of the object ($j = 1$) and surrounding fluid ($j = 2$), respectively. For diamagnetic materials $\chi < 0$, while for paramagnetic materials $\chi > 0$. For a vertical-axis solenoid magnet, a stable equilibrium point, corresponding to a local minimum in E_{MG} , is possible for objects with $\chi_1 < \chi_2$ and $D_1 > D_2$; the conditions are discussed in detail by Berry and Geim [43]. This minimum, if it exists, lies on the solenoid axis above the center of the solenoid, where $\partial B / \partial z < 0$. Less well known is the possibility of stable levitation of objects with $D_1 < D_2$, such as a bubble of gas in a liquid, provided that $\chi_1 < \chi_2$. In this case, stable levitation occurs *below* the center of the solenoid, where $\partial B / \partial z > 0$. Studies by Pichevant *et al.* [50], Duplat and Mailfert [51], and more recently Mongruel *et al.* [52] have pioneered the use of this method to study bubbles of oxygen gas levitated in liquid oxygen at cryogenic temperatures. Magnetically levitated foams have also been studied [53] in which the experiments were conducted in the upper region of the solenoid. Here we use this technique to study (at room temperature) an isolated air bubble in water to which a small amount of a paramagnetic salt has been added. Air bubbles levitated this way are not significantly deformed by the magnetic field, unlike bubbles in liquid oxygen, as we show. The bubble is trapped in stable mechanical equilibrium at a particular point in the magnetic field, away from the container walls.

II. EXPERIMENTAL METHOD

We used a custom-built superconducting magnet (Cryogenic Ltd., London) to levitate air bubbles in a weakly paramagnetic aqueous solution of manganese chloride (0.18 mol L^{-1} , $\chi_2 = 2.5 \times 10^{-5}$). This magnet has previously been used to study, for example, free liquid drops [54], granular and fluid systems under differential gravity [47,55] and combined acoustic and magnetic levitation [56]. The magnet has a room-temperature, 58 mm-diameter cylindrical bore. A 1 L cylindrical tank of external diameter 57 mm was filled with the manganese chloride solution and inserted into the bore as shown in Fig. 1. The top and bottom of the tank were fitted with acrylic windows. The interior of the tank was illuminated through the bottom window by a 0.4 W white LED. A 45-degree mirror was placed above the tank allowing the bubble to be imaged through the top window using a high-speed camera (Phantom Miro M340) positioned approximately 2 m away from the magnet. A frame rate of up to 5000 frames per second was used with a resolution of 20–30 μm per pixel.

The strength of the magnetic field was 10.6 T at the center of the solenoid. For this magnet at this field strength, the location of the potential minimum in E_{MG} , the point where the bubble is in stable equilibrium, lies 99 mm below the center of the solenoid, labeled L_0 in the contour plot of E_{MG} in Fig. 1. In the vicinity of L_0 , the magnetogravitational force $-\nabla E_{\text{MG}}$ acts as a restoring force towards this point, increasing in strength approximately linearly with distance for small displacements. This force provides an additional weak cohesive force on the bubble, in addition to surface tension, and therefore, the frequency of shape oscillations is expected to increase under its influence. Our calculations (Sec. SII [29]) show this effect to be very small, however: we expect an increase in the normal mode frequencies of approximately 0.2% for bubbles of the size studied in these experiments (see also [57,58]).

Since the potential E_{MG} is not precisely spherically symmetric about L_0 , as can be seen from Fig. 1(a), some deformation of the equilibrium shape of the levitated bubble away from spherical is also expected. This deformation only becomes significant if the surface tension, which acts to minimize the surface area, is comparable to or less than

$$\gamma_c \approx \frac{1}{8\mu_0} \left| \frac{3\chi}{2} R^3 \left[\left(\frac{\partial B}{\partial z} \right)^2 + 2B \frac{\partial^2 B}{\partial z^2} \right] - \frac{\chi}{3} R^4 \frac{\partial^3 B^2}{\partial z^3} \right|, \quad (9)$$

adapting a calculation by Duplat and Mailfert [51] (see Sec. SIII [29]). Here the magnetic field strength B and its derivatives are to be evaluated at the levitation point L_0 . For a bubble of radius 2.9 mm levitating in the trap shown in Fig. 1(a), Eq. (9) gives $\gamma_c \sim 0.1 \text{ mN m}^{-1}$, much less than the

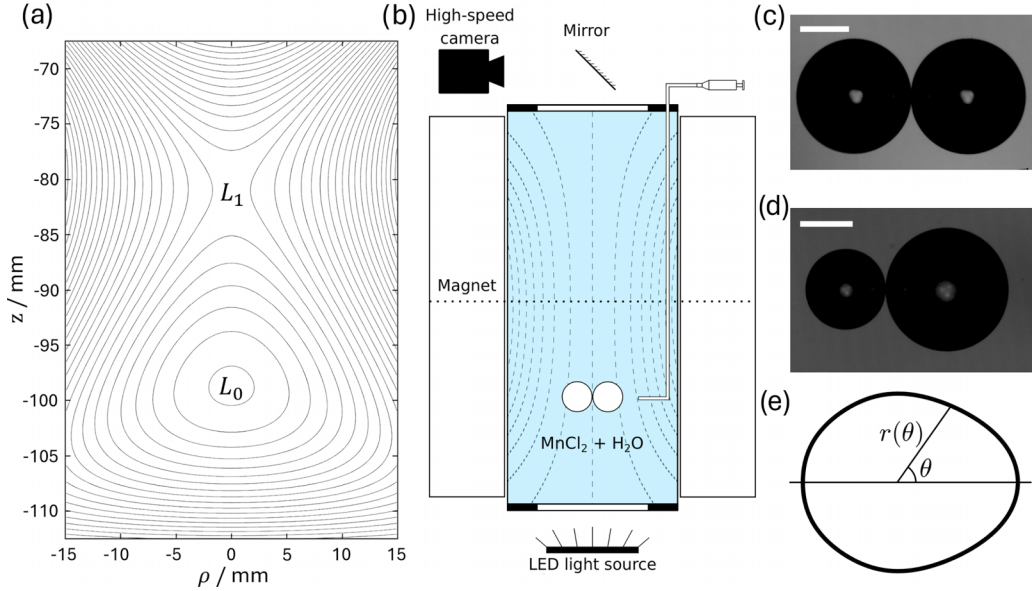


FIG. 1. (a) Contour plot of the magnetogravitational potential, E_{MG} , of an air bubble in water containing dissolved manganese chloride (0.18 mol L^{-1}). The cylindrical coordinates ρ (radial) and z (vertical) are measured from the geometric center of the solenoid coil. L_0 and L_1 mark points of stable and unstable equilibrium, respectively. (b) Schematic of the experimental setup (not to scale), showing the liquid-filled tank inserted into the magnet bore. Two bubbles are injected into the liquid at L_0 using an air-filled syringe connected to a stainless steel tube. The bubbles are viewed and illuminated through windows fitted in the top and bottom of the tank. Curved dashed lines represent magnetic field lines. Images of the parent bubbles, viewed from the top of the tank, immediately before coalescence are shown in (c) and (d) for the symmetric case and asymmetric case, respectively. White scale bars represent 2 mm. (e) Schematic illustrating the bubble shape (after coalescence) represented in polar coordinates $r(\theta)$.

actual surface tension of the bubble, $\gamma \approx 70 \text{ mN m}^{-1}$, and so we expect the equilibrium shape of the bubble to be close to spherical. Indeed, numerical solution of the Young-Laplace equation [59] shows that the radius of the equilibrium shape is expected to vary by, at most, 0.05% over the surface of the bubble (Sec. SIII [29]), that is, by a maximum of $1.5 \mu\text{m}$.

A prolate distortion, elongating the bubble in the direction of the magnetic field, is also expected as a consequence of forces proportional to χ^2 arising from the demagnetizing field of the bubble in the paramagnetic liquid, as is apparent in experiments on levitating bubbles of oxygen gas in liquid oxygen [51]. However, since the susceptibility of the room temperature manganese chloride solution used in these experiments is approximately 140 times smaller than that of liquid oxygen at its boiling point, the distortion is expected to be relatively weak here; we estimate a variation in the radius of up to 0.01% over the bubble's surface by this effect.

We injected two air bubbles at the stable levitation point L_0 using a stainless steel tube (1.0 mm inner diameter) connected at one end to a 10 ml air-filled plastic syringe. The other end of the tube was inserted into the liquid as shown in Fig. 1(b), so that it was positioned 5–10 mm away from L_0 . An air bubble was produced by slowly pushing the syringe plunger, which detached from the end of the tube once it reached a critical size determined by the balance of surface tension and the magnetogravitational force $-\nabla E_{MG}$. Adjusting the distance between the tube's open end and L_0 altered the magnitude of the magnetogravitational force acting on the emerging bubble, allowing us to control the diameter of the released bubble. The second bubble was produced by repeating the process. Since the shape of the equipotential surfaces of E_{MG} close to L_0 in these experiments is slightly oblate [see Fig. 1(a)] the two bubbles came to rest aligned horizontally either side of L_0 . The

TABLE I. Radii of the initial two parent bubbles at rest, R_1 and R_2 . Radius at rest, R , and the capillary time, τ , of the bubble formed from the coalescence.

Case	R_1 (mm)	R_2 (mm)	R_2/R_1	R (mm)	τ (ms)
Symmetric	2.31 ± 0.03	2.31 ± 0.03	1.00 ± 0.02	2.90 ± 0.03	19.18 ± 0.01
Asymmetric	1.60 ± 0.02	2.46 ± 0.02	1.54 ± 0.02	2.66 ± 0.02	16.60 ± 0.01

parent bubbles remained in this state, pushed weakly into contact by the magnetogravitational force, for 3–4 seconds before the onset of coalescence. Figures 1(c) and 1(d) show the initial condition for the symmetric and asymmetric cases respectively. The radii of each of the parent bubbles are given in Table I, along with the capillary time, τ , and radius, R , of the bubble formed from their coalescence. The capillary time was measured directly from the frequency of small-amplitude oscillations of the $n = 2$ mode of the bubble, $\tau = \sqrt{12}/\omega_2^{(0)}$.

From each video frame we extracted the surface profile of the bubble using the Sobel edge detection method [60]. The surface contour of the bubble was measured in polar coordinates, $r(\theta, t)$, where θ is the polar angle measured from the axis passing through the centres of the original two spherical parent bubbles [Fig. 1(e)], and t is the time measured from the onset of coalescence. We then decomposed $r(\theta, t)$ as a series of surface harmonics [3]: $r(\theta, t) = \sum_{l=0} s_l(\theta, t)$. This requires $r(\theta)$ to be single valued for all θ , which is true except for very early times after coalescence and for a brief period during the pinch-off of a small satellite bubble early in the coalescence. Since the coalescence is expected to be axisymmetric about this axis, we assume that the surface harmonics may be written purely in terms of the zonal harmonics $s_l(\theta, t) = A_l(t)P_l(\theta)$. The origin of the polar coordinate system (r, θ) is chosen such that $A_1 = 0$.

III. NUMERICAL SIMULATIONS

The incompressible Navier-Stokes equations were solved for both liquid and gas phases using the open source multiphase volume of fluid solver Basilisk [61,62]. An axisymmetric domain was used to reduce computational cost. The simulated meridional plane was taken to be a square with side lengths equal to $12R_s$, where R_s is the radius of the smallest of the two parent bubbles. This relatively large domain size was used to minimize wall effects on the simulation. Free-slip boundary conditions were enforced at the three external edges of the domain and symmetry conditions were enforced along the central symmetry axis of the domain. Time steps were determined using a Courant-Friedrichs-Lewy number of 0.5. Further information on the time stepping and advection schemes is given in Refs. [61] and [63]. A quadtree dynamic adaptive mesh refinement (AMR) scheme [61] was used to refine the mesh resolution at every time step at the liquid-gas interface and in areas of relatively large velocity, down to a minimum cell side length of $0.0059R_s$. Grid adaption is based on minimizing the error estimated using a wavelet algorithm; further information on this algorithm may be found in Popinet [62]. The AMR scheme constrained the maximum side length of any cell at the interface to be $0.0059R_s$ for all time steps. The high local resolution was required to accurately model the capillary-driven flows, particularly in the region of the neck formed between the two parent bubbles immediately after coalescence. An example of one of the meshes used is shown in the Supplemental Material (Sec. SIV [29]). Surface harmonics were obtained from simulations, determining the surface contour $r(\theta, t)$ at intervals of $t = 0.01\tau$, following the same procedure used for the experimental images.

IV. RESULTS

A series of images showing the oscillations of a bubble formed from the coalescence of two equal-sized parent bubbles (symmetric case) and two parent bubbles with radii in the ratio 1:1.54 (asymmetric case) are shown in Figs. 2 and 3, respectively. The overlaid contours show the shapes

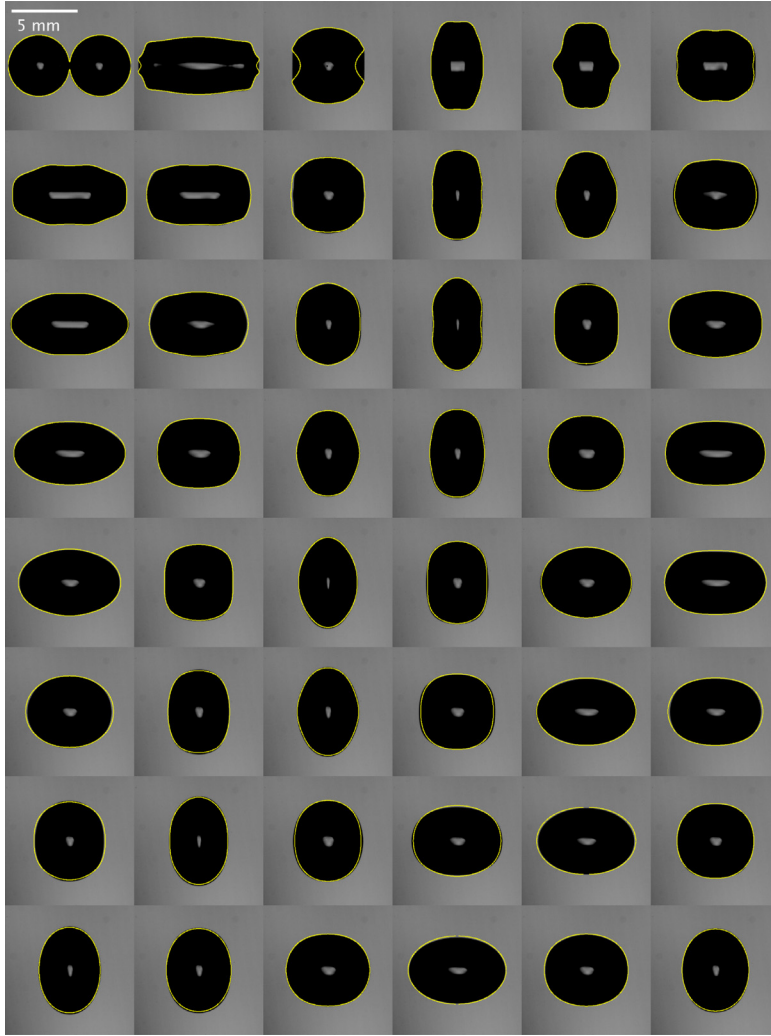


FIG. 2. Series of images showing the oscillations of a magnetically levitated air bubble in water, formed from the coalescence of two same-sized parent bubbles (first frame). Time increases from left to right and then from top to bottom, with an interval between each frame of 6.8 ms (0.35τ). Yellow contours show the shapes obtained from numerical simulation.

calculated by the numerical simulations for comparison. Differences arise between simulation and experiment directly after coalescence when the shapes of the bubbles are reentrant, since the concave features are obscured from the camera's view by the foreground bubble surface, but otherwise the experiments and simulations agree well. In experiment, the parent bubbles were observed to remain at rest for a short time after their injection into the liquid, as the fluid drained from the small gap between them. Once started, coalescence proceeded with the rapid growth of a neck generating axisymmetric capillary waves that traveled around each bubble away from the neck towards the opposite poles of the parent bubbles, as previously observed [64,65]. The capillary waves formed protrusions at these poles which then pinched off to form small satellite bubbles in both cases. From the images, it can be observed that there are contributions from several surface harmonics in the subsequent oscillations, in both symmetric and asymmetric cases. In the symmetric case, the motion consists predominantly of two-lobed oscillations with contributions from shapes with

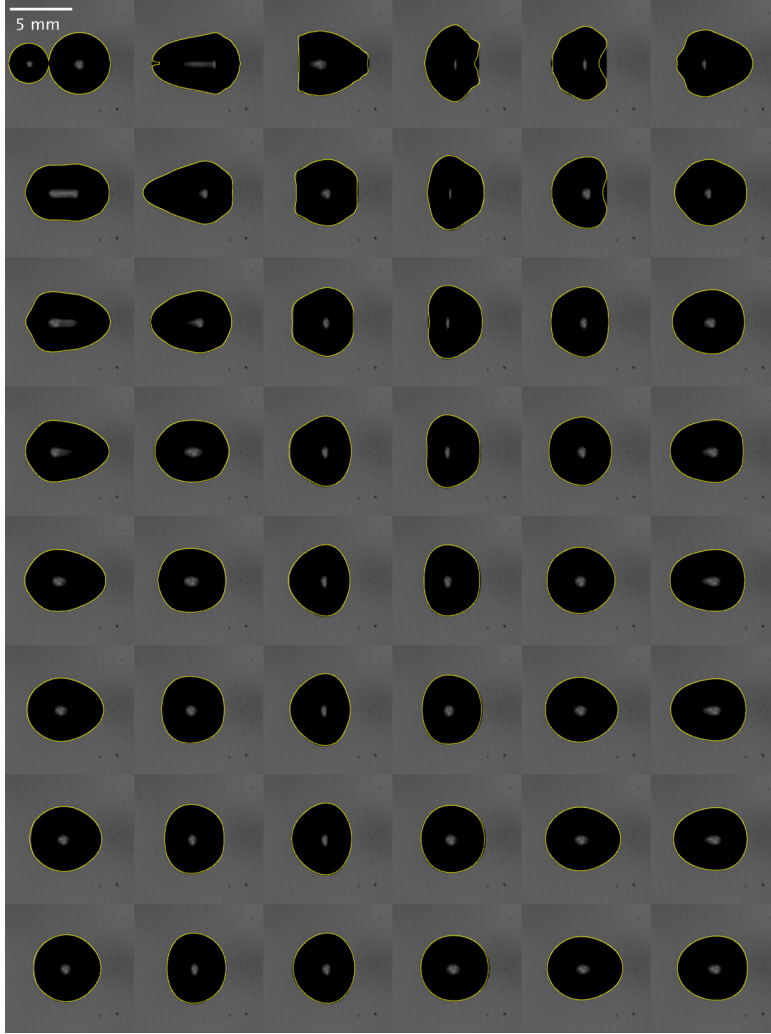


FIG. 3. Series of images showing the oscillations of a magnetically levitated air bubble in water, formed from the coalescence of two parent bubbles with radii in the ratio 1:1.54 (first frame). Time increases from left to right and then from top to bottom, with an interval between each frame of 5.5 ms (0.33τ). Yellow contours show the shapes obtained from numerical simulation.

a four-lobed contour at early times. In the asymmetric case, a shape oscillation with a three-lobed contour is also evident. Figure 4 shows the oscillations in time of the surface harmonics, $A_l(t)$, normalized by the radius of the spherical bubble at rest, R , up to $l = 6$. Each surface harmonic oscillates at multiple frequencies and decays with time due to viscous damping, with the higher degree harmonics decaying more quickly. In the symmetric case, no oscillations in the odd-degree harmonics were observed in simulation as expected, since the odd-degree surface harmonics are asymmetric about $\theta = \pi/2$. In experiment, oscillations of small amplitude $A_l < 0.05$ were observed in the $l = 3$ and $l = 5$ surface harmonics in this case (Sec. SV [29]), which are due to a small asymmetry between the parent bubbles, with a difference in radius smaller than the uncertainty in its measurement. Apart from an obvious difference in the $l = 0$ oscillations, discussed later, there is generally good agreement between the A_l obtained in experiment and simulation.

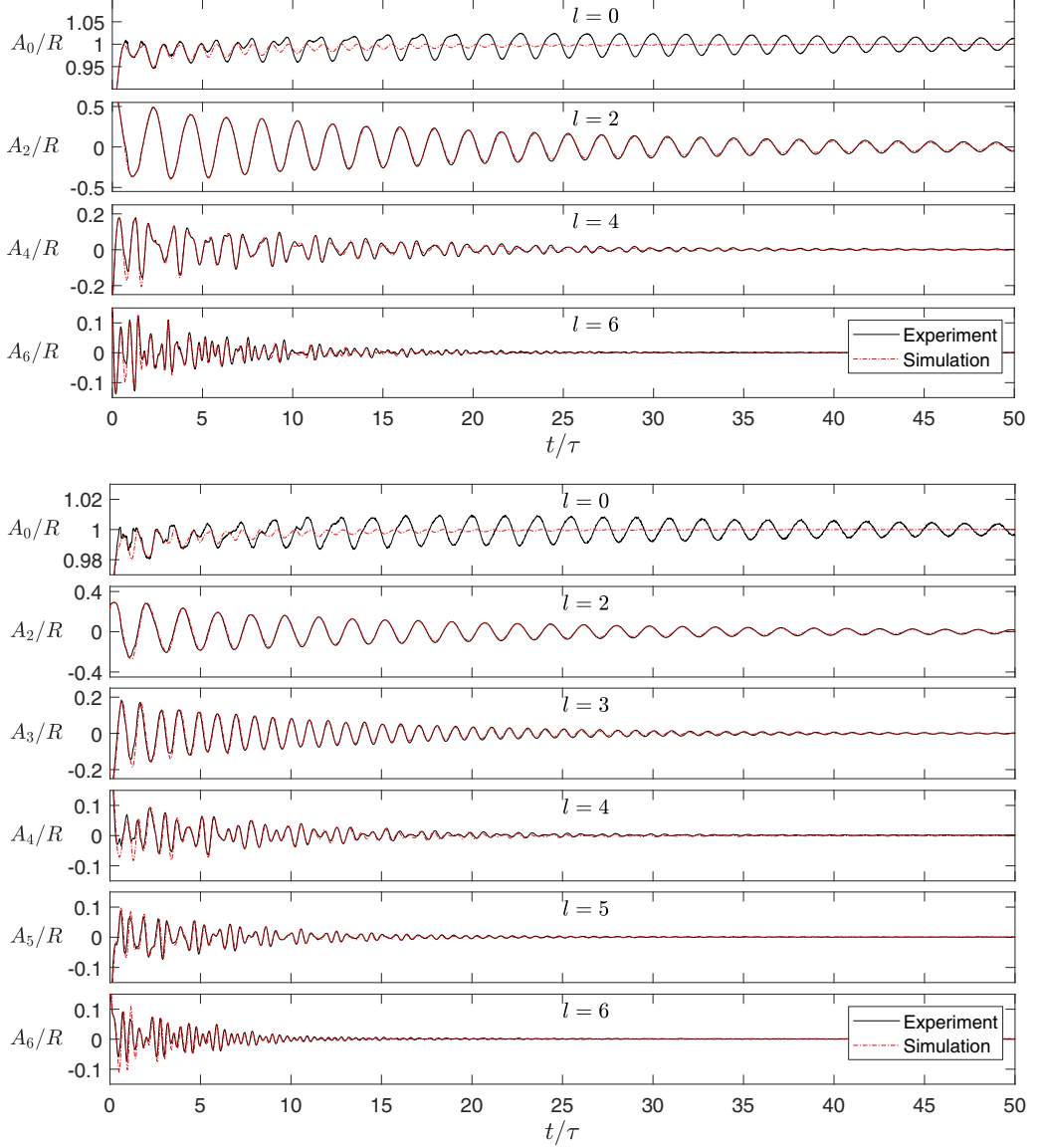


FIG. 4. Oscillations in time of the surface harmonics $A_l(t)/R$ of degree $l = 0-6$ for the symmetric (a) and asymmetric case (b), where R is the radius of the spherical bubble at rest. In (a), only oscillations of harmonics of even degree are displayed since the A_l of odd degree harmonics are small in experiment and zero in simulation. A_1 is omitted since the origin of the coordinate system is chosen such that $A_1 = 0$. Note that the scales of the vertical axes differ for each harmonic.

The spectrum of the oscillations of each surface harmonic was obtained from the short-time Fourier transform (STFT):

$$y(\omega, t) \equiv \mathcal{F}[x(t')H(t' - t)]. \quad (10)$$

Here $x(t)$ is the input signal, $x(t) = A_l(t)/R$ for the surface harmonics $2 \leq l \leq 6$, and $x(t) = A_0(t)/R - 1$ for the $l = 0$ surface harmonic; $\mathcal{F}[X(t)]$ is the discrete Fourier transform of $X(t)$ and $H(t)$ is the Hann window function, of duration 16τ . Figure 5 shows the linear spectra obtained

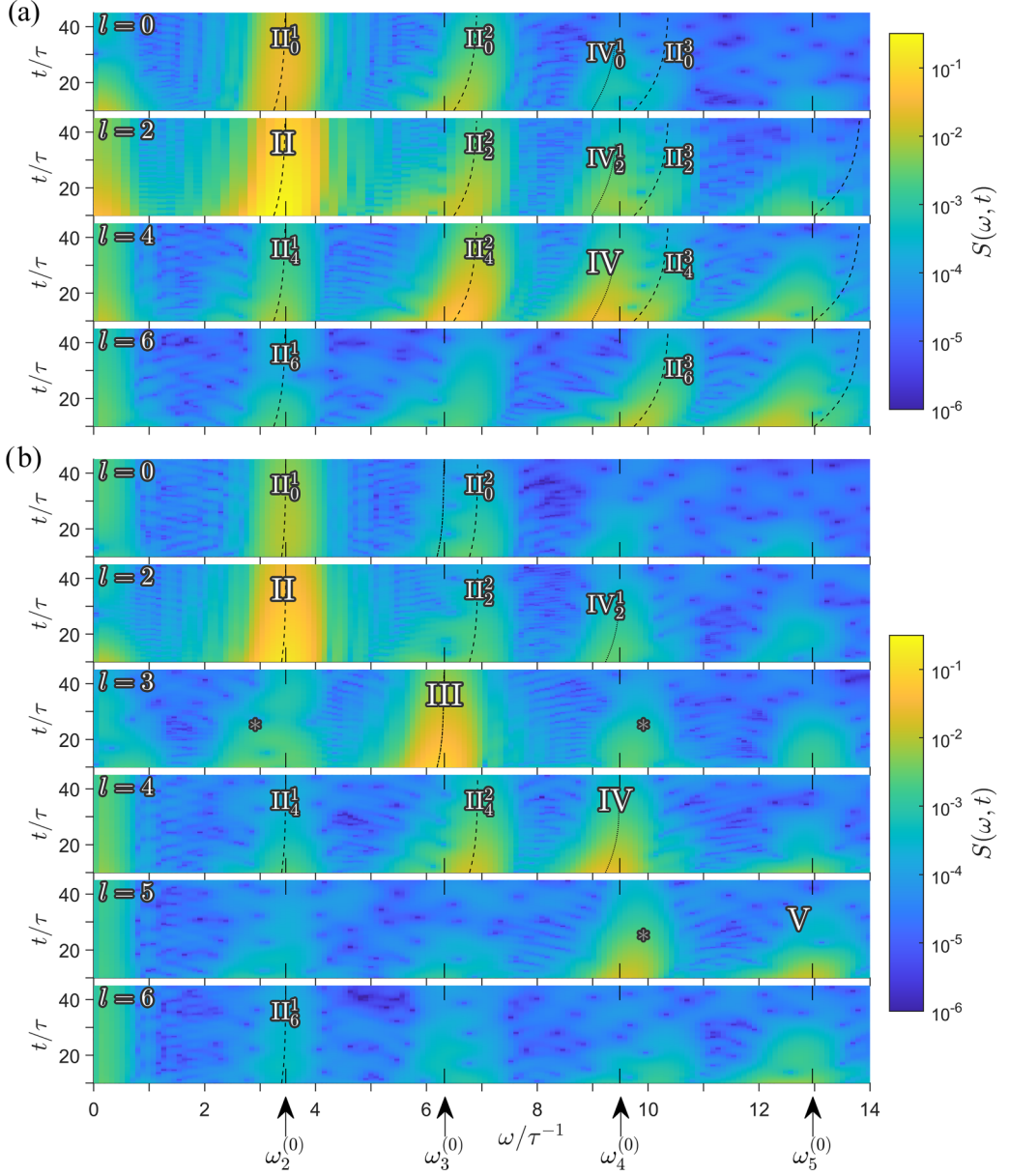


FIG. 5. Linear spectra $S(\omega, t)$ obtained from the short-time Fourier transform of the oscillations of the surface harmonics, $A_l(t)/R$, $l \leq 6$, in the symmetric (a) and asymmetric (b) experiments. Color indicates the value of S on the logarithmic scale shown by the color bars, right. Peaks in the spectra appear as bright yellow and green stripes. The broken curved lines indicate the frequencies of the prominent peaks labeled II (dash), III (dot-dash), and IV (dotted)—that is, the frequency at peak maximum—and also integer multiples of these. The arrows on the x axis point to additional tick marks indicating the frequencies of the linear modes of oscillation, $\omega_n^{(0)}$. The $l = 1$ spectrum is omitted since the origin of the coordinate system is chosen such that $A_1 = 0$.

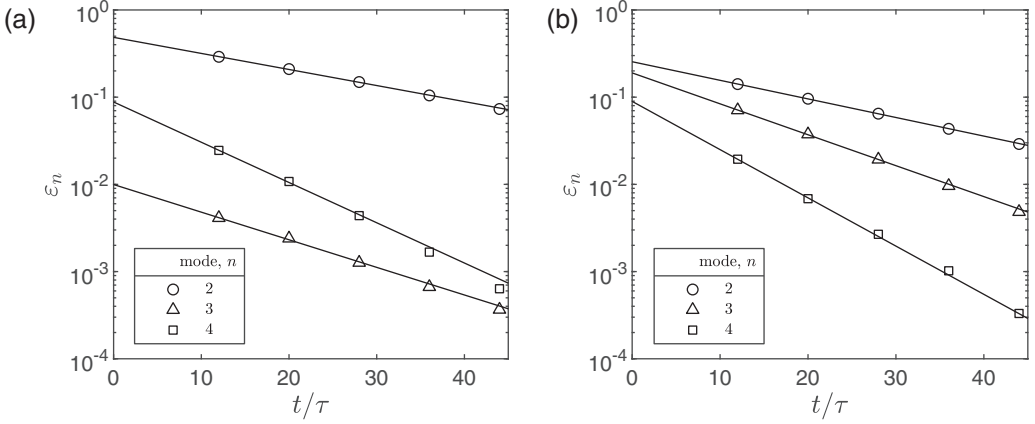


FIG. 6. Measured amplitudes ε_n of the two-, three-, and four-lobed modes ($n = 2$ –4) as a function of time t after the start of the coalescence, in the symmetric (a) and asymmetric (b) experiments. The lines are fits to the data with the model $\varepsilon_n(t) = \varepsilon_n(0)e^{-\alpha_n t/\tau}$ (see main text). The values of the fitting parameters $\varepsilon_n(0)$ and α_n are given in Table II.

from the magnitude of the STFT, $S(\omega, t) \sim |y(\omega, t)|$ (Sec. SVI [29]; see also [66]). The height of each peak in $S(\omega, t)$ gives the amplitude of the input signal at that frequency and time. For example, a purely sinusoidal input, $x = a \sin \omega t$, produces a peak in S with height a at frequency ω . Further details of the calculation of the linear spectrum from the STFT are given in the Supplemental Material (Sec. SVI [29]). We have shown only the spectra from experiment here since the spectra obtained from the simulations do not reveal any obvious differences, except in the case of $l = 0$, where the peak at $\omega \approx \omega_2^{(0)}$ visible in the $l = 0$ spectrum is much weaker in simulation. Both data sets are shown in the Supplemental Material (Sec. SVII [29]). Prominent peaks, labeled II, III, IV, etc., are visible at frequencies ω_n close to $\omega_n^{(0)}$ in the spectra of the oscillations of the surface harmonics of degree $l = n$. These peaks correspond to the first-order contribution to the bubble shape, $\varepsilon P_n(\theta) \cos(\omega_n t)$ [Eqs. (2) and (3)]. We thus measure the frequency ω_n and the amplitude ε_n of the corresponding modes from the frequency and height, respectively, of these peaks. The broken curved lines in Fig. 5 show the measured frequencies $\omega_n(t)$ of each of these peaks, which increase with time, approaching $\omega_n^{(0)}$ asymptotically. We also plot the same curves, $\omega_n(t)$, as dashed lines overlaying the spectra of other surface harmonics in Fig. 5, and also integer multiples of ω_n , to aid in identifying the other peaks visible in the spectra. Peaks at ω_2 , $2\omega_2$, and $3\omega_2$ are clearly visible in the spectra of the $l = 0, 2, 4$, and 6 surface harmonics in the symmetric case, for example. We label these peaks, which correspond to contributions to the bubble shape at higher order in ε , with a Roman numeral according to the oscillation mode, a subscript l indicating the shape harmonic, and a superscript p indicating the frequency multiple.

Figure 6 shows the variation in time of the amplitudes ε_n of the two-, three-, and four-lobed modes in experiment, obtained from the heights of peaks II, III, and IV (Fig. 5): $\varepsilon_2 = S(\text{II})$, $\varepsilon_3 = S(\text{III})$ and $\varepsilon_4 = S(\text{IV})$. The data were sampled at time intervals of 8τ . The plots show the amplitudes decaying exponentially with time. Similar data were obtained from simulation. We extracted the initial amplitudes, $\varepsilon_n(0)$, and decay constants, α_n , of these modes by fitting the model $\varepsilon_n(t) = \varepsilon_n(0)e^{-\alpha_n t/\tau}$ to the experimental data (Table II). The initial amplitude of the two-lobed mode, $\varepsilon_2(0)$, is significantly larger in the symmetric case than in the asymmetric case. The initial amplitude of the four-lobed mode, $\varepsilon_4(0)$, is similar in both cases. In the symmetric case, we expect the amplitude of the three-lobed mode, ε_3 , to be zero. In experiment, however, a small peak is visible at this frequency (Sec. SVII [29]), giving an initial amplitude $\varepsilon_3(0) = 0.01$, which we attribute to a small asymmetry between the parent bubbles, as previously mentioned. Decay constants are similar in both coalescences.

TABLE II. Initial amplitudes $\varepsilon_n(0)$ and decay constants α_n of the two-, three-, and four-lobed modes ($n = 2, 3, 4$), obtained by fitting the model $\varepsilon_n(t) = \varepsilon_n(0)e^{-\alpha_n t/\tau}$ to the experimental data shown in Fig. 6. Values in parentheses represent 95% confidence intervals.

n	Parameter	Symmetric	Asymmetric
2	$\varepsilon_n(0) \times 10^3$	485 (465, 505)	255 (251, 259)
	$\alpha_n \times 10^4$	424 (406, 442)	491 (483, 499)
3	$\varepsilon_n(0) \times 10^3$	10 (9, 11)	190 (180, 200)
	$\alpha_n \times 10^4$	730 (660, 800)	815 (780, 850)
4	$\varepsilon_n(0) \times 10^3$	90 (80, 100)	89.5 (82.0, 97.0)
	$\alpha_n \times 10^4$	1060 (990, 1130)	1275 (1210, 1340)

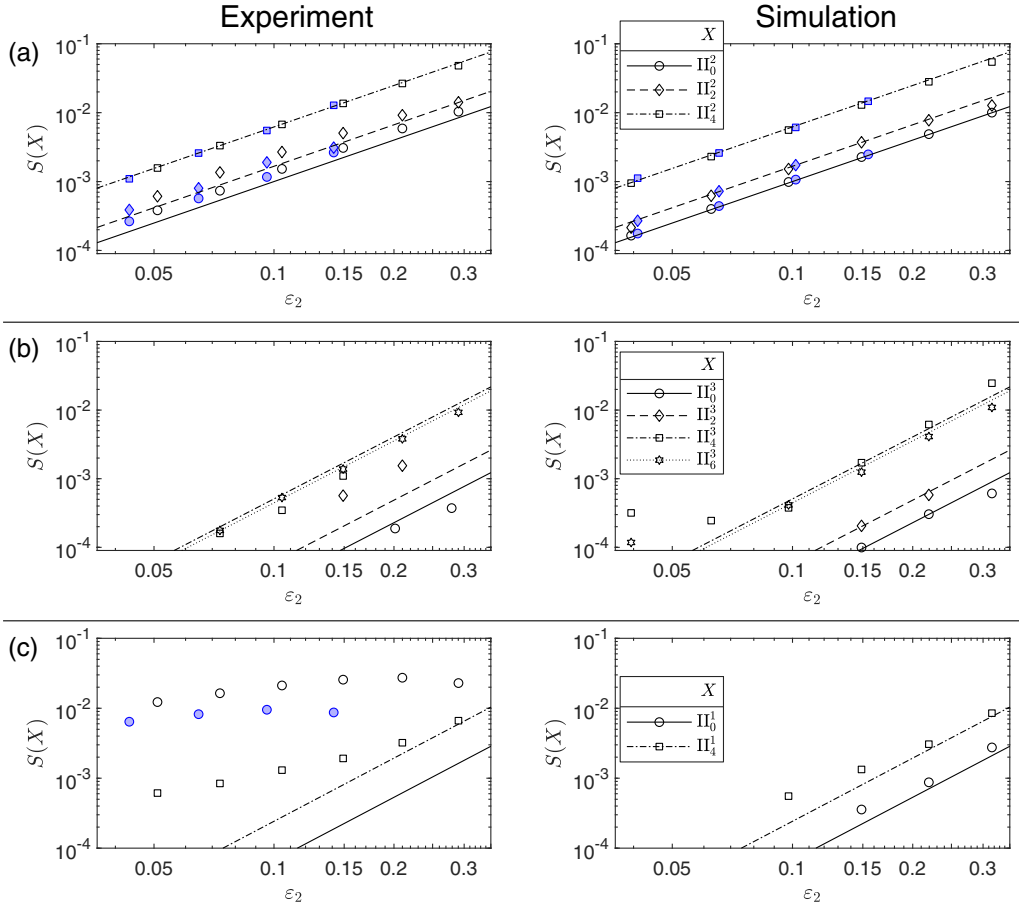


FIG. 7. Log-log plots of the heights of higher order peaks Π_l^p in the spectra plotted against the measured amplitude ε_2 of the two-lobed mode. The peaks are grouped according to their frequencies in the spectra: (a) $2\omega_2$, (b) $3\omega_2$, and (c) ω_2 . Open black and solid blue markers show data obtained from the symmetric and asymmetric coalescences, respectively. Lines $S \propto \varepsilon_2^2$ in (a) show the expected heights of the second order peaks according to the theory of T&B. Lines $S \propto \varepsilon_2^3$ plotted in (b) and (c) show the expected heights of the third-order peaks according to our calculation given in the Supplemental Material, Sec. SI [29].

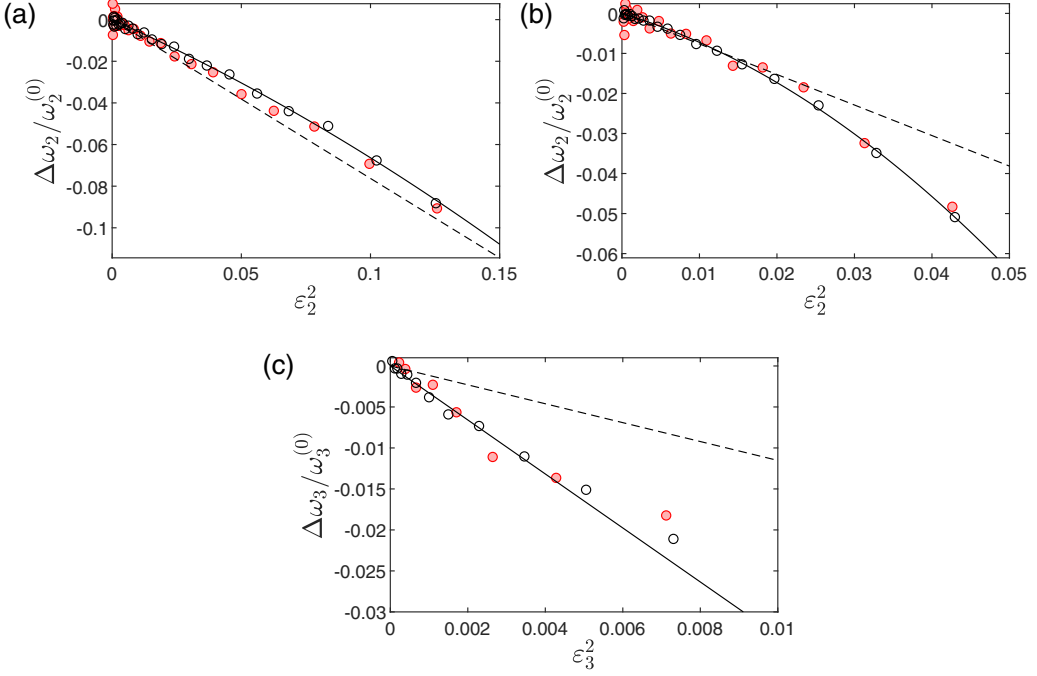


FIG. 8. Relative shift in the frequency of the two-lobed oscillations ($n = 2$), $\Delta\omega_2/\omega_2^{(0)}$, as a function of the square of its amplitude, ε_2^2 , for symmetric (a) and asymmetric (b) cases; and relative shift in frequency of the three-lobed oscillations ($n = 3$), $\Delta\omega_3/\omega_3^{(0)}$, as a function of ε_3^2 , for the asymmetric case (c). Hollow black and solid red markers show data from experiment and simulation, respectively. Broken lines show the shift predicted by T&B to $O(\varepsilon^2)$ in both cases. Solid lines show fits to the experimental data with the model $\Delta\omega_2/\omega_2^{(0)} = a\varepsilon_2^2 + b\varepsilon_2^4$ in the case of $n = 2$ and $\Delta\omega_3/\omega_3^{(0)} = a\varepsilon_3^2$ in the case of $n = 3$. The values of the fitting parameters a and b are given in Table III.

Figure 7 shows the heights $S(\Pi_l^p)$ of the higher order peaks Π_l^p in the spectra, as a function of the measured amplitude of the two-lobed mode, $\varepsilon_2 = S(\Pi)$. We focus our analysis of the higher order peaks on this set because they are the clearest and most consistently resolved in the spectra. The peaks are grouped according to their frequencies: Fig. 7(a) shows peaks with frequency $2\omega_2$, and Figs. 7(b) and 7(c) show peaks with frequency $3\omega_2$ and ω_2 . Figure 7(a) includes measurements from both symmetric (open symbols) and asymmetric cases (solid symbols). Figures 7(b) and 7(c) show mostly data for the symmetric case since the corresponding peaks in the asymmetric case become too weak to obtain an accurate measurement of their height as the oscillations decay. Peak Π_6^1 is too weak to measure accurately in either case. The lines show the heights of the peaks expected from (4) and (5) for comparison, discussed further in Sec. V below.

Figures 8(a) and 8(b) show the relative change in frequency, $\Delta\omega_n/\omega_n^{(0)}$, of the two- and three-lobed modes as a function of their measured amplitudes ε_2 and ε_3 , obtained from the frequencies and heights of peaks II and III in the spectra. Since the data are sampled at equal time intervals (at half the width of the Hann window function), and since the amplitudes ε_n decay exponentially with time, the density of data points decreases with increasing ε_n in the plot. Broken lines show the expected relative change in ω obtained by T&B (7) to second order in ε , i.e., $\Delta\omega_n/\omega_n^{(0)} = \varepsilon^2\omega_n^{(2)}/2\omega_n^{(0)}$. Solid lines represent fits of the experimental data using $\Delta\omega_2/\omega_2^{(0)} = a\varepsilon_2^2 + b\varepsilon_2^4$ for the two-lobed mode and $\Delta\omega_3/\omega_3^{(0)} = a\varepsilon_3^2$ for the three-lobed mode [67]. The measured values of the second- and fourth-order frequency coefficients obtained from the fits to both experimental and simulation data, $\omega_n^{(2)}/\omega_n^{(0)} = 2a$ and $\omega_n^{(4)}/\omega_n^{(0)} = 24b$, are given in Table III.

TABLE III. Measured values of the second- and fourth-order frequency coefficients, $\omega_n^{(2)}/\omega_n^{(0)} = 2a$ and $\omega_n^{(4)}/\omega_n^{(0)} = 24b$, obtained from the fits to the data shown in Fig. 8, using the model $\Delta\omega_2/\omega_2^{(0)} = a\varepsilon_2^2 + b\varepsilon_2^4$ for the two-lobed oscillation ($n = 2$) and $\Delta\omega_3/\omega_3^{(0)} = a\varepsilon_3^2$ for the three-lobed oscillation ($n = 3$). Values in parentheses represent 95% confidence intervals.

Experiment			
n	Case	$-\frac{\omega_n^{(2)}}{\omega_n^{(0)}}$	$-\frac{\omega_n^{(4)}}{\omega_n^{(0)}}$
2	Symmetric	1.11 (1.04, 1.18)	27 (20, 34)
2	Asymmetric	1.17 (1.08, 1.26)	340 (300, 370)
3	Asymmetric	6.6 (6.3, 6.9)	N/A
Simulation			
2	Symmetric	1.33 (1.18, 1.48)	7 (−10, 25)
2	Asymmetric	1.09 (0.70, 1.48)	330 (220, 440)
3	Asymmetric	6.4 (6.0, 6.8)	N/A

In the asymmetric case, but not in the symmetric case, we also observed motion of the center of mass of the bubble in the direction of the smaller of the two parent bubbles. Figure 9 shows the displacement of the center of mass along the symmetry axis of the bubble as a function of time, in both experiment and simulation.

V. DISCUSSION

In general there is good agreement between the results obtained from experiment and simulation, as is evident from the time series of the surface harmonics $A_l(t)$ shown in Fig. 4. However, differences arise where the simulations calculate a surface contour that is concave at the “poles” of the bubble, as seen in the third image in Fig. 2, for example. In experiment, these concavities are hidden from view of the camera by the surrounding bubble surface, producing discrepancies between experiment and simulation in the oscillations of the surface harmonics. These are only significant at early times $t \lesssim 3\tau$ and do not affect the spectra shown in Fig. 5 and the data obtained from them. The oscillations of the $l = 0$ harmonic are an obvious exception: while the results from experiment and simulation initially agree, a growing oscillation appears at ω_2 in $A_0(t)$ in both the symmetric and asymmetric experiments, while the oscillations decay in simulation [68]. We discuss the origin of this divergence below.

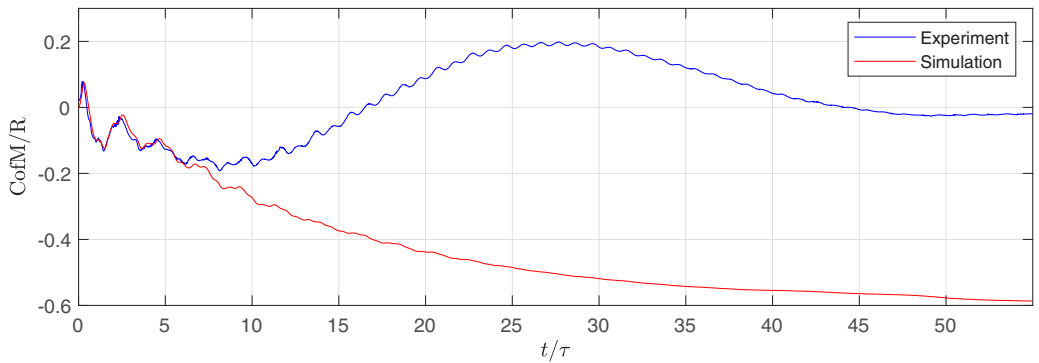


FIG. 9. Displacement of the center of mass (CofM) of the bubble along its axis of symmetry as a function of time, for the asymmetric case.

Consistent with our prediction (Sec. II) we did not detect any deviation from spherical of the equilibrium shape of bubbles levitated by the magnetic field. This was true even for bubbles levitated side by side (the initial state in these experiments) where the magnetogravitational force pushes the two bubbles together, toward L_0 . Even in this case, where we expect the equilibrium shape of the bubbles to show the largest distortion, we were unable to measure any distortion of the equilibrium shape within the uncertainty given by the resolution of these images. For the two equal-sized bubbles shown in Fig. 1(c), for example, the ratio of the bubble radii measured along two axes, along and perpendicular to the line joining the centres of the two bubbles, was 0.997 ± 0.004 .

We now compare our results with T&B's calculations for the shape oscillations of a bubble to second order in amplitude, and with our calculations of the shape oscillations to third order in amplitude. As discussed in the introduction, T&B's solution for each n -lobed oscillation mode consists of a surface harmonic of degree $l = n$ oscillating at frequency ω_n (3), with smaller contributions (by a factor ε) from surface harmonics of degree $l = 0, 2, \dots, 2n$ oscillating at $2\omega_n$ (4) [69]. We observe these components as peaks in the frequency spectra of the surface harmonics (Fig. 5). Peak II, appearing in the $l = 2$ harmonic with a frequency close to $\omega_2^{(0)}$, corresponds to the linear term in the expansion of the shape function (2) for this mode, $\varepsilon P_2(\theta) \cos(\omega_2 t)$ (3). We therefore measure the amplitude $\varepsilon_2(t)$ and frequency $\omega_2(t)$ of the two-lobed mode from the height and frequency of this peak. Similarly, peaks III, IV, and V, near $\omega_3^{(0)}$, $\omega_4^{(0)}$, and $\omega_5^{(0)}$, represent the linear contributions to the shapes of the three-, four-, and five-lobed oscillations ($n = 3, 4, 5$), respectively, enabling the measurement of their respective amplitudes ε_n and frequencies ω_n .

Peaks Π_0^2 , Π_2^2 , and Π_4^2 , appearing in the $l = 0, 2$, and 4 harmonics, have frequencies twice that of peak II. These three peaks correspond to the second-order contributions to the shape for this mode. This is confirmed by the log-log plots of the heights of these peaks against the measured amplitude of the mode, $\varepsilon_2 = S(\Pi)$, shown in Fig. 7(a). Lines show the heights of these peaks expected from the amplitudes of the oscillating second-order terms in the shape function [Eqs. (2) and (4)], $S(\Pi_l^2) = |\frac{1}{2}\varepsilon_2^2 c_{2,l}|$, using the coefficients $c_{2,l}$ obtained by T&B: $c_{2,0} = -\frac{1}{5}$, $c_{2,2} = -\frac{1}{3}$ and $c_{2,4} = \frac{304}{245}$ [1]. The measured peak heights agree well with those predicted, and particularly well in simulation.

Including corrections to the shape at third order in ε (Sec. SI [29]) adds contributions from surface harmonics $l = 0, 2, \dots, 3n$ for even n and $l = 1, 3, \dots, 3n$ for odd n , oscillating at ω_n and $3\omega_n$ (5). The two sets of peaks Π_l^1 and Π_l^3 that are visible in the spectra correspond to these contributions to the two-lobed mode. The lines in Figs. 7(b) and 7(c) show the expected heights of these peaks as a function of ε_2 : $S(\Pi_l^1) = |\frac{1}{6}\varepsilon_2^3 e_{2,l}(1 + f_{2,l})|$ and $S(\Pi_l^3) = |\frac{1}{6}\varepsilon_2^3 e_{2,l}f_{2,l}|$, with coefficients $e_{2,l} = -\frac{4}{7}, \frac{501}{1372}, \frac{662122}{147147}, -\frac{114562}{31691}$ and $f_{2,l} = -\frac{3}{10}, -1, -\frac{2241551}{3310610}, -\frac{4141605}{5613538}$ for $l = 0, 2, 4, 6$, respectively (Sec. SI [29]). We have omitted the line for Π_6^1 since the peak heights measured in simulation and experiment are too weak to measure accurately. Since $f_{2,2} = -1$, there is no contribution to $l = 2$ at ω_2 expected at $O(\varepsilon^3)$; that is, we expect $S(\Pi_2^1) = 0$. There is generally good agreement between the simulation results and theory. In experiment, peaks Π_0^3 , Π_4^3 , and Π_6^3 are also in good agreement with theory. The proximity in frequency of this set of peaks, Π_l^3 , to those at ω_4 hinders their resolution, however, potentially contributing to the observed discrepancy between theoretical and experimental peak heights for Π_2^3 . Peaks Π_0^1 and Π_4^1 in experiment exhibit quite different behavior from that expected from the theory and are discussed further below. There are other visible peaks, for which the measurements of their peak heights are too noisy to identify unambiguously. For example, peaks at approximately $\omega = 4\omega_2$ are visible in the spectra, suggesting that they correspond to fourth-order corrections to the shape.

Figure 8 shows the change in the frequency of the two- and three-lobed oscillations as a function of their amplitudes ε_2 and ε_3 , obtained from the frequency and height of peaks II and III in the spectra. In the symmetrical experiment and simulation, the frequency of the two-lobed oscillations varies approximately quadratically with amplitude ε as expected from the T&B theory. However, the asymmetric coalescence data show the presence of a significant quartic term (ε^4), evident in the plot (Fig. 8) and quantified by the coefficient $\omega_2^{(4)}$ given in Table III. Though the quartic contribution to

the frequency shift of the two-lobed mode differs significantly between symmetric and asymmetric coalescences, the quadratic contribution ($\omega_2^{(2)}$) is similar in both cases (Table III). The values of $\omega_2^{(2)}/\omega_2^{(0)}$ obtained from experiment and simulation broadly agree with each other (within their respective uncertainties), pointing to a value of $\omega_2^{(2)}/\omega_2^{(0)} \approx -1.1$ to -1.3 , inconsistent with T&B's result, -1.526 [70,71]. We found no explanation for this discrepancy in our method or analysis. The consistency between simulation and experiment suggests that the discrepancy is not specific to the methods used in either. An error in measuring the amplitude of the mode from the peak II height that could account for the difference would show in the plot of the heights of the second-order peaks $S(\Pi_7^2)$ versus that of the first-order peak $S(\Pi)$ (Fig. 7) as a clear deviation from the theoretical prediction, whereas we observe excellent agreement with theory. Although viscous damping results in a correction to the free decay frequency, this is expected to be insignificant for the bubble sizes considered here, of the order 0.1% of $\omega_n^{(0)}$ [34], assuming that the viscosity of the solution is the same as that of water.

For the three-lobed oscillations observed in the asymmetric coalescence, the measured value of $\omega_3^{(2)}/\omega_3^{(0)} = 2a \approx -6.5$ obtained from both the asymmetric experiments and simulation peak III data is considerably larger in magnitude than the predicted value of -2.305 .

The symmetric and asymmetric cases are distinguished by the amplitudes of the modes that are excited by each coalescence. The oscillations resulting from the symmetric coalescence are dominated by the contribution from the two-lobed shape mode. The mode with the next largest amplitude is the four-lobed mode, an order of magnitude smaller in amplitude. The contribution of the three-lobed mode is small in experiment ($\varepsilon_3(t=0) = 0.01$) and zero (below the noise) in simulation, due to symmetry in this case. The ratio of the amplitudes of the two- and four-lobed modes is approximately $\varepsilon_4/\varepsilon_2 \approx 0.18$ at $t=0$ dropping to $\varepsilon_4/\varepsilon_2 \approx 0.05$ at $t=20\tau$ as the higher frequency $n=4$ oscillations damp more quickly. Thus, the coalescence of two equal-sized bubbles produces oscillations that decay quickly to a good approximation of the “moderate” amplitude two-lobed oscillations considered by T&B. In contrast, the asymmetric coalescence generates oscillations with comparable contributions from two- and three-lobed oscillations: the ratio of their amplitudes are $\varepsilon_3/\varepsilon_2 \approx 0.74$ at $t=0$, dropping to $\varepsilon_3/\varepsilon_2 \approx 0.38$ by $t=20\tau$. Interaction of the two-lobed oscillation with the three-lobed oscillation is thus responsible for the apparent ε^4 dependence of $\Delta\omega_2$ in the asymmetric case but not the symmetric case. This interaction may also explain the anomalous value of $\omega_3^{(2)}$. We note that a similar discrepancy was reported by Kornek *et al.* [37] for nonlinear oscillations of soap-film bubbles, where two- and three-lobed oscillations were also excited with comparable amplitudes. While the three-lobed oscillation is significant only in the asymmetric coalescence, the four-lobed oscillation, though relatively weak, is present with equal amplitude in both symmetric and asymmetric cases. This suggests that its coupling with the two-lobe mode may explain the consistently lower than predicted value of $\omega_2^{(2)}$ observed in both cases.

The peaks marked by asterisks, which appear in the spectra of the $l=3$ and $l=5$ harmonics in the asymmetric coalescence (as shown in Fig. 5), are related to the oscillatory motion observed in the bubble's center of mass (Fig. 9). Specifically, these peaks correspond to the components of the center of mass oscillation at frequencies close to $\omega_2^{(0)}$ and $\omega_4^{(0)}$. As the oscillations decay, these peaks shift to higher frequency, approaching values just below $\omega_2^{(0)}$ and just above $\omega_4^{(0)}$. A number of studies have shown how such center of mass motion can be generated from the coupling of modes of adjacent degree [36,72,73]. The divergence after $t \approx 6\tau$ of the center of mass measured in experiment from that in simulation can be explained by the influence of the magnetogravitational force, which provides a retarding force increasing in magnitude as the bubble moves away from the equilibrium position L_0 , eventually reversing its direction at $t \approx 10\tau$.

Finally, we return to the observed discrepancy between simulation and experiment in the $l=0$ oscillations $A_0(t)$. The spectrum of $l=0$ oscillations shows a corresponding peak Π_0^1 in the experimental data at ω_2 , which is an order of magnitude larger than is observed in the simulation data [Fig. 7(c)] and does not vary with amplitude as ε^3 as expected. The same behavior can be

reproduced in simulation by rotating the three-dimensional model of the bubble approximately 10 degrees away from horizontal and then repeating the polar decomposition of the three-dimensional bubble shape projected onto the horizontal plane, mimicking the imaging process in experiment. The height of peak Π_4^1 is also increased significantly by doing this, reproducing the larger than expected height of this peak observed in Fig. 7(c) in the experimental data. The other peaks are either unaffected by the rotation, or show only relatively minor changes. Thus, despite the parent bubbles being horizontally aligned at the onset of coalescence as shown in Fig. 1, our results suggest a small rotation of the symmetry axis away from horizontal as they coalesce.

VI. SUMMARY AND CONCLUSION

The nonlinear axisymmetric shape oscillations of 5–6-mm-diameter air bubbles in water, produced through coalescence, were studied using magnetic levitation and compared with results from both numerical simulation and theory. Analysis of the spherical surface harmonics of the oscillations showed that coalescence of two equal-sized parent bubbles produced a single bubble undergoing “moderate–large”-amplitude oscillations dominated by a single two-lobed mode ($n = 2$) of oscillation, approximating the idealized situation considered by T&B, whereas coalescence of two unequal-sized parent bubbles, differing in diameter by a factor of 1.54, produced oscillations with comparable contributions from both two- and three-lobed ($n = 2$ and 3) oscillations. The good agreement between simulation and experiment, aside from a difference in the $l = 0$ oscillations explainable by a small change in the angle of the symmetry axis in experiment, validates the simulation results and indicates the absence of significant experimental artefacts. The oscillation frequency ω_2 of the two-lobed mode varies with its amplitude ε_2 as $\Delta\omega_2 = \frac{1}{2}\omega_2^{(2)}\varepsilon_2^2 + O(\varepsilon_2^4)$ as predicted by T&B, though our measurements of the second-order frequency component $\omega_2^{(2)}$ in both simulation and experiment, and in both symmetric and asymmetric cases, range from $\omega_2^{(2)}/\omega_2^{(0)} = -1.1$ to -1.3 , compared to the predicted value of -1.526 . In the asymmetric coalescence, there is also a significant contribution to $\Delta\omega_2$ at ε^4 . Our measurement of the second-order frequency component of the three-lobed mode observed in the asymmetric coalescence, $\omega_3^{(2)}/\omega_3^{(0)} \approx -6.5$ is larger in magnitude by a factor of 2.8 compared to the theoretical prediction, -2.305 , in both experiment and simulation. We attribute this difference, and the observation of the significant ε^4 component of the frequency shift in the asymmetric coalescence, to coupling between the two- and three-lobed modes excited with comparable amplitudes in this case.

The measured amplitudes of the surface harmonic oscillations associated with the two-lobed mode closely match T&B’s second-order predictions that were obtained for motion consisting purely of two-lobed shape oscillations. This agreement holds not only for the symmetric coalescence, in which the contributions to the resulting oscillations from other modes are small, but also for the asymmetric coalescence which involves comparable contributions from both two- and three-lobed oscillations. Furthermore, for the symmetric coalescence, the measured amplitudes of the surface harmonics of the two-lobed mode agree well with our calculations at third order. Measurement noise prevented verification of this agreement in the asymmetric coalescence.

These experiments demonstrate that magnetic levitation allows one to study bubbles of air in water, singly and in pairs, of relatively large size (a few millimeters in diameter), trapped in stable mechanical equilibrium. An important advantage of this method of levitation over other methods is that there is negligible distortion of the equilibrium shape of the air bubble by the forces used to levitate it. These features, we believe, make magnetic levitation attractive for use more widely, as an alternative “microgravity” platform for studying bubble phenomena.

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DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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