A method for triggering surface modes by bubble coalescence

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Abstract

Acoustically driven microbubbles find numerous applications in the biomedical field (targeted drug delivery or blood-brain barrier opening for instance) and in engineering (surface cleaning or sonochemistry). In most cases only spherically oscillating bubbles are considered in these techniques. However, bubbles exhibiting surface modes can lead to additional effects (enhanced micromixing amongst others), and are scarcely investigated (mainly due to positional and growing surface instabilities). We present an experimental setup which allows the triggering and control of surface modes of free bubbles. As opposed to earlier studies which are based on short-time exposure to ultrasound waves [1] or an amplitude modulated pressure field [2], we obtain bubbles with steady-state surface modes. The experimental procedure consists of the following steps: (i) An acoustic field is induced in a water tank by a transducer. The driving frequency is chosen as to create an acoustic standing wave inside the tank. (ii) A bubble is nucleated by a short focused laser pulse. Our bubbles are all below resonance size and are hence driven towards a pressure antinode due to primary Bjerknes forces. (iii) By nucleating a second bubble, the two bubbles will coalesce due to primary Bjerknes forces and mutual attraction by secondary Bjerknes forces. The coalescence leads to a single, initially deformed bubble. By choosing the appropriate set of acoustic pressure and bubble size, a desired surface mode can be triggered. Additionally, we observed that the trajectory of the coalescing bubbles will define the orientation of the axis of symmetry. In sum, coalescence is a fast method to induce surface modes in free acoustically driven bubbles.

Keywords: microbubbles; shape modes; surface modes; bubble coalescence

Introduction

Shape modes of microbubbles have mainly been investigated theoretically. First theoretical studies were based on uncoupled mode theory [3,4]. These models allow in particular predicting the appearance of parametrically excited surface modes of order *n* when the driving amplitude exceeds a certain pressure threshold p_n^{thresh} . Experimentally, the onset surface modes can be observed by exposing bubbles to short ultrasound bursts [1].
Coupling between different modes has been introduced with studies on energy transfer between the spherical mode and one or two shape modes [5,6]. Later, models were extended to include all energy transfer between translational, spherical and surface modes [7,8]. These models allow taking into account the saturation of the shape mode growth. They are hence able to describe stead-state dynamics of a bubble with surface modes. Experimental validations of these models have been effected [2] using amplitude-modulated sound fields.
The above cited experimental methods of exposure to short ultrasound bursts and the amplitude-modulated sound fields allow the triggering and subsequent analysis of temporal dynamics. Steady-state oscillations of surface modes are however difficult to obtain as their stability has to be assured. We propose bubble coalescence as a means of triggering surface modes.

Experimental setup

The experimental setup is presented in Figure 1. It consists of a 8cm-edge cubic tank filled with water. On the bottom area, an ultrasonic plane transducer (SinapTec®, diameter of the active area 35mm) is connected to it in order to create an acoustic field inside the tank. The driving frequency is set to 31.25kHz and the voltage amplitude of the transducer can be varied between 0 and 10V. No gain amplifier is used as the estimated acoustic pressure in the tank for 5V applied voltage is ~30kPa, a sufficient value to generate surface modes for nearly all our experimentally studied bubbles. Backlight illumination with a continuous light source (LED) and a CMOS camera (Vision Research® V12.1) equipped with a 12x objective lens (Navitar®) allow capturing the bubble dynamics. A best compromise between frame size and frame rate is found to be 256x256 pixels and 67.065 kHz. Single bubbles are nucleated by focused laser pulses (λ= 532nm, New Wave Solo III, 6 ns pulse duration). All nucleated as well as coalesced bubbles are smaller than 90µm. To capture coalescence, a first bubble is trapped in the acoustic field, then a second bubble will be nucleated and encounter the first one. An exemplary result is shown in Figure 2. In order to simplify the manipulation during the experiments, the position of the water tank can be adjusted by millimeter screws in all three directions.

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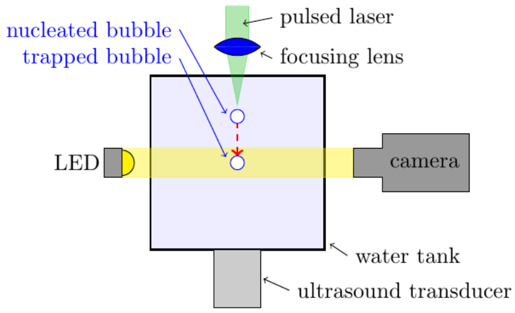

Figure 1: Schematic presentation of the experimental setup.

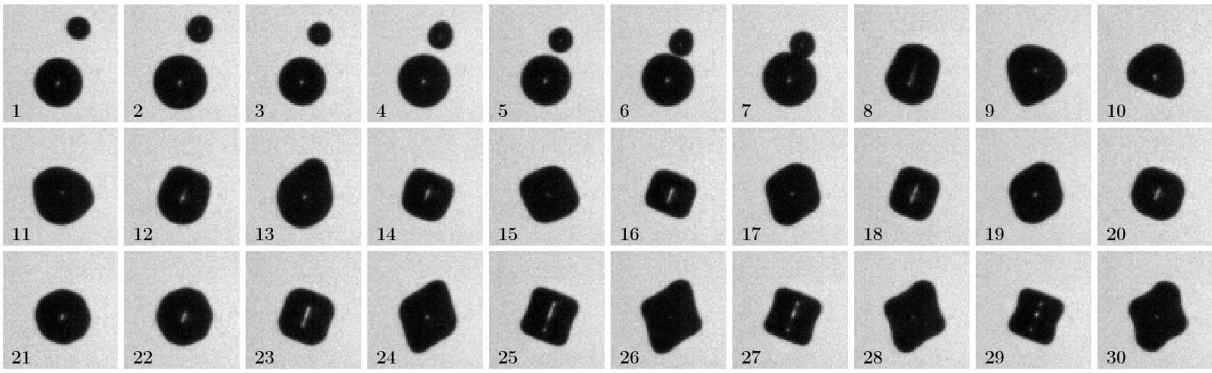

Figure 2: Exemplary series of 30 consecutive snapshots of two coalescing bubbles resulting in a shape mode 4. The images are recorded at time intervals of 14.9µs. The initial bubble radii are 52.5µm and 24.2µm respectively, the acoustic pressure has been evaluated to be 24.5kPa.

A microbubble in an acoustic field

A gas bubble in water, exposed to an acoustic field will be influenced by several forces. The bubble oscillations caused by the pressure fluctuations has been approached by different theoretical investigation, one of them being the Keller-Miksis model [12]. Fitting the Keller-Miksis model to the experimentally obtained bubble radius allows deducing the acoustic pressure on the bubble if there is no direct pressure measurement.
The bubble position is defined by the sum of forces acting on the bubble. Those are mainly the force of buoyancy and the primary Bjerknes forces. Primary Bjerknes forces are the net forces $\langle F \rangle = -\langle V \nabla p \rangle$ induced by the pressure gradients ∇p of the acoustic field onto the bubble volume V [13]. In a relatively weak acoustic field (hence omitting large bubble oscillations leading to translational instability and so-called dancing motion [14]), bubbles smaller than resonant size oscillate in-phase with the acoustic field, bubbles larger than resonant size out-of-phase. Consequently, the former are driven towards a pressure antinode, the latter are driven towards a pressure node. According to Minnaert's theory [15], the resonant radius corresponding to 31.25kHz is R_{res}=104µm. All our studied bubbles are hence smaller than resonant size and driven towards a pressure antinode. The bubble will find a stable position closely above a pressure antinode to equal out the buoyancy. Hence, the exact position depends on the bubble size. Here, the importance to have a rapidly adjustable tank position becomes obvious.
As soon as a second bubble is nucleated, there are also mutual forces between the two bubbles, called secondary Bjerknes forces. For bubbles oscillating in-phase with one another, secondary Bjerknes forces cause attraction between them. These forces will therefore generally cause the first bubble to leave its equilibrium location before encountering the second one. The coalesced bubble will later find a new equilibrium location slightly beneath the original one.

Stability of surface modes

There are two main difficulties, when we wish to obtain bubbles with stable surface modes. They are (i) the triggering of the surface mode and (ii) the stability of the surface mode as well as the appearance of surface modes alone without any translational instability. No studies in literature present a method which overcomes all of those difficulties. Exposing bubbles to short ultrasound bursts (see e.g. [1]) allows triggering surface modes. Also, due to the short pulse duration the shape mode amplitude and the possible appearance of translational instabilities can be limited. However, no steady-state regime can be obtained. Another existing method is to drive bubbles with an amplitude-modulated ultrasound field (see e.g. [2]), where the driving frequency is modulated with a low modulation frequency. This allows periodic triggering of non-spherical oscillations and their observation during short time intervals (before their extinction) that will obviously limit their growth. However, this technique does not allow for a steady-state regime. It is also possible to place a bubble in an acoustic field and rise the acoustic pressure until surface modes appear. However, without an additional triggering, bubbles often rest in a metastable spherical mode before abruptly instabilities take over. With very careful tuning, stable shape modes can be obtained in this way. But as this method is laborious and does not always succeed, it is desirable to find another method to trigger surface modes. Bubble coalescence can be used to do so.

Conditions for triggering surface modes

A surface mode can be triggered if the acoustic pressure exceeds the respective pressure threshold. This threshold $p_n^{thresh}(f_{osc},R_o)$ of a mode of order n depends on the driving frequency f_{osc} and the bubble radius at rest R_o [4]. Numerical calculations of the stability regions for a fixed driving frequency f_{osc}=31.25kHz are given in Figure 3. For a chosen set of parameters, the triggered mode can hence be predicted. For example, for a coalesced bubble with a radius of 70µm at 10kPa we expect to obtain a mode 3, whereas for the same bubble at 5kPa we expect to obtain a purely spherically oscillating bubble. Figure 4 shows three exemplary series of bubble coalescence with similar sized bubbles but different acoustic pressures resulting respectively in a spherical mode, a mode 2 and sustained rebound. Sustained rebound of bubbles has been observed in some cases (see Figure 4c)). The exact conditions under which the bubbles do not coalesce have however not yet been studied in detail. In literature, one of the discussed parameters is the Weber number [9-11].

The map in Figure 3 allows choosing the correct parameters in order to trigger a desired mode. We can slightly influence the size of the nucleated bubble by tuning the power of the pulsed laser and we can grow the trapped bubble by multiple coalescences. The acoustic pressure amplitude is directly related to the transducer voltage. In our study, a linear relation between the pressure and the voltage has been found in preliminary experiments. During these experiments, a spherically oscillating bubble has been driven at different transducer voltages. By fitting a Keller-Miksis model [12] to the experimentally obtained radii, the respective pressures can be deduced.

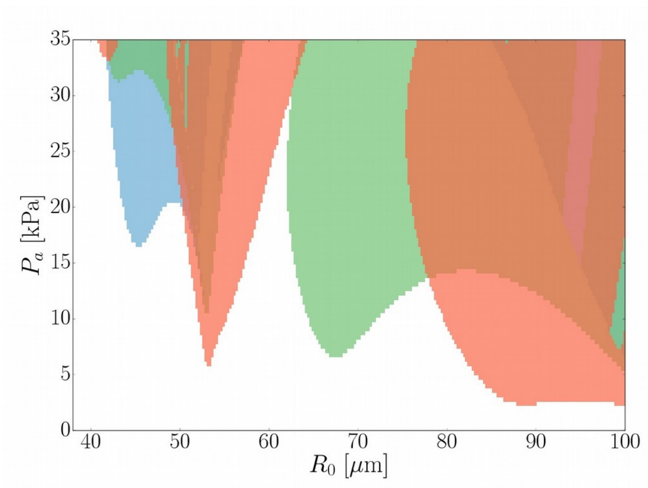

Figure 3: Radius-pressure map for a driving frequency f_{osc}=31.25kHz giving the stability boundaries and indicating the theoretically expected mode for bubble radii R_0=[40µm 100µm] and driving pressures P_a=[0kPa 35kPa]. White – spherical mode, blue – mode 2, green – mode 3, red – mode 4. Theory based on [4].

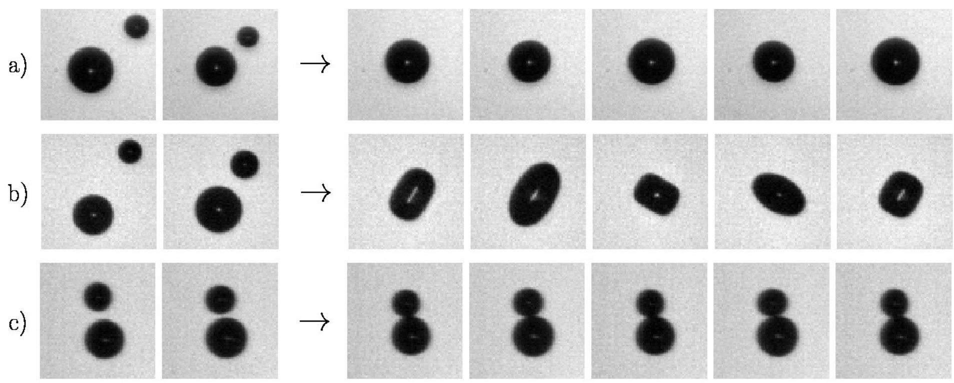

Figure 4: Three examples of similar sized bubbles at different acoustic pressures resulting in different shape modes: a) 40.9µm and a 21.8µm radius bubble at 27kPa coalescing to a spherically oscillating bubble b) a 40.8µm and a 24.3µm bubble 28kPa resulting in a shape mode 2 c) a 36.9µm and a 25.9µm radius bubble at 13kPa resulting in sustained rebound. For all series the first two snapshots are taken at about 0.3s and 0.1s before the encounter of the two bubbles, the following images are five consecutive snapshots taken at about 1s after the encounter.

The axis of symmetry

Nearly all experimental and theoretical studies on shape modes of bubbles are based on the expansion of the bubble interface as a sum of Legendre Polynomials (meaning zonal harmonics amongst the basis of spherical harmonics). This approach supposes axisymmetry which seems to be fairly correct for a large number of observed bubbles. However, for experimental purposes, the definition of the axis of symmetry is crucial for a correct post-processing of the bubble shape. The easiest way to post-process without any additional correction and without loss of information requires the axis of symmetry to be in the plane of the camera.
Earlier studies have shown that the axis of symmetry can be defined by the direction of traveling ultrasound waves [1]. However, in the present setup no such possibility is given as we work with a standing wave without any preferential direction. To achieve the definition of the axis of symmetry, our experiments show that coalescence is a means to influence it. It is defined by the direction of approach between the two bubbles. An example is demonstrated in Figure 5. Earlier classical studies on bubble coalescence (e.g. [10]) already let foresee these hypothesis by presenting coalescing ellipsoidal bubbles respecting the here formalized symmetry axis. With the present study we are able to formalize this and in particular to validate its extension to surface modes of higher order.

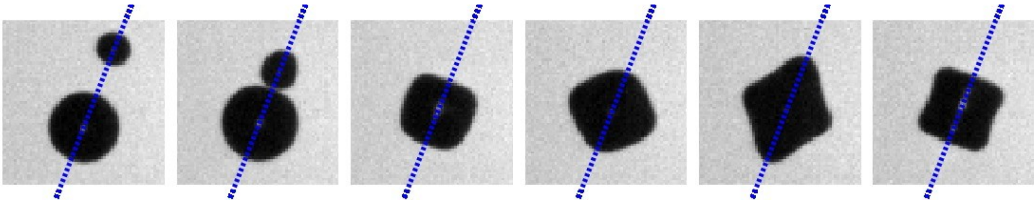

Fig 5: Demonstration of the axis of symmetry for some chosen snapshots of Figure 3.

Conclusion

We present bubble coalescence as a reliable method to trigger surface modes. As opposed to earlier methods such as short exposition to ultrasound pulses or modulated pressure fields, the coalesced bubbles can remain in a steady-state regime. With the definition of the axis of symmetry, it is possible to completely control the appearing shape mode, including the shape mode number and the orientation of the bubble. In conclusion bubble coalescence is a fast method to trigger surface modes employable for further investigation of bubble dynamics.

Acknowledgements

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