

Acoustic pulsation of a microbubble confined between elastic walls

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This paper reports an experimental and theoretical study of the dynamics of microbubbles flattened between the two walls of a microfluidic channel. Using a micropit, a single bubble is trapped by capillarity at a specific position in the channel and its oscillation under ultrasound is observed by stroboscopy. It is shown that the bubble dynamics can be described by a two-dimensional Rayleigh-Plesset equation including the deformation of the walls of the channel and that the bubble behaves as a secondary source of Rayleigh waves at the wall interface. Above a critical pressure threshold, the bubble exhibits a two-dimensional shape oscillation around its periphery with a period doubling characteristic of a parametric instability. We report how each shape mode appears, varying the bubble radius and the amplitude of excitation, and demonstrate that the wall deformation has no significant effect on their dynamics. © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4942917]

I. INTRODUCTION

The acoustic properties of air bubbles in water have received much attention from both experimental and theoretical points of view, whether for the energy that bubbles can release by cavitation¹ or for their utility as ultrasound contrast agents in medicine.² This interest for bubble dynamics is motivated by the particularly strong response of the bubble to insonication.³ For low ultrasound intensities, the bubble undergoes an axisymmetric vibration, also called breathing mode, and develops an additional shape instability above a certain intensity threshold of the insonifying field.⁴ These oscillations are favored by the large difference in acoustic impedance between air and water that leads to a strong coupling with the external field.

Three-dimensional (3D) microbubbles attached to a channel wall have already proved their efficiency in sonoporation applications:^{5,6} the bubble pulsation induces a transient membrane permeabilization of biological cells, allowing the introduction of genes into the cells without the help of a virus. More recently, Ahmed *et al.* also showed that horseshoe-trapped microbubbles confined in a two-dimensional (2D) channel were very promising for mixing applications in low Reynolds environments.⁷ Therefore, the potential of confined microbubbles could be of huge interest in both medical and industrial applications. To the best of our knowledge, an appropriate model describing the dynamics of confined microbubbles is still needed for a better control of these microfluidic processes.

In this paper, we propose a detailed study of the dynamics of a microbubble squeezed between the two walls of a microfluidic channel. Because this bubble is strongly flattened in this microfluidic environment, it appears as quasi-2D, meaning that its oscillating motion mainly occurs in the plane of the channel. In order to fix the position of the bubble inside the channel, we have developed a novel approach inspired by the method of Dangla *et al.*⁸ to handle droplets. This allowed the study of the bubble dynamics under controlled acoustic conditions.

In the first part, we report that the axisymmetric pulsation of such a bubble has a resonance shifted to the low frequencies, compared to a spherical bubble of the same radius. We propose

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FIG. 1. Experimental setup: Sketch of the microfluidic channel (height $h = 25 \ \mu$ m, width $w = 2 \ m$ m), where a pancake-like bubble (blue) is excited by the ultrasonic waves generated by the piezoelectric plate glued on the glass bar (green). The liquid flows along e_x^* . Zoom window: Expanded view of the bubble (radius $R_0 = 15-75 \ \mu$ m) anchored on its cylindrical micropit (diameter $d = 20-35 \ \mu$ m).

a model based on Rayleigh-Plesset equation, taking into account the deformation of the channel walls. With this model, we demonstrate that the bubble in fact pulsates as if it was in a small cylinder of liquid. The characteristic size of this cylinder is determined by the wavelength of the Rayleigh waves generated by the bubble pulsation. In a second part, we show that for high intensities of ultrasound (\sim 30–100 kPa), the acoustic field does not only drive an axisymmetric oscillation of this quasi-2D bubble, but can also trigger a *parametric instability* leading to surface mode vibration. Because the channel is thinner than the wavelength of the surface modes, these modes are two-dimensional and develop only in the plane of the channel. The amplitude of the modes and their rotation are analyzed. A phase diagram of each surface mode is sketched, highlighting the characteristic resonance tongues predicted by the Mathieu equation. Furthermore, we demonstrate that the wall deformation has no significant impact on the resonance of the surface mode.

II. MATERIALS AND METHODS

A. Experimental setup

The experimental setup consists in a microfluidic device made of two layers of polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning) where the PDMS elastomer is mixed with the cross-linker in a weight ratio of 10:1 and cured at 65 °C for 3 h. The upper layer is composed of a flow focusing orifice⁹ connected to a rectangular main channel (height $h = 25 \ \mu$ m, width $w = 2 \ \text{mm}$), as described in Ref. 10. To introduce ultrasound in the channel, a piezoelectric plate is glued on a glass bar. This glass bar (4 × 40 mm) is encased into the upper PDMS layer, about 150 μ m above the main channel. Because the wavelength of the sound is of same order as the thickness of the bar, the Lamb modes¹¹ of the glass bar are excited and a standing wave develops on the glass surface.

A micropit (40 μ m in height, 20–35 μ m in diameter) is patterned on the lower layer¹² (see Fig. 1). The two layers are then bonded together by an oxygen plasma treatment (Harrick Plasma), placing the micropit in front of the glass bar. This geometry is an improvement of the device used in Ref. 10 since the presence of the pit allows to study bubbles either in motion or at rest at a specific position in the channel.

B. Methods

Using the flow focusing orifice, monodisperse bubbles are continuously produced by forcing nitrogen into a solution of water containing 5% of a commercial dishwashing detergent (Dreft, Procter and Gamble). The surfactant was added to prevent undesired dewetting of the bubbles. By controlling the air inlet pressure and the liquid flow rate, we could obtain confined bubbles of various volumes, with radii R_0 between 15 and 75 μ m corresponding to a bubble shape ranging from a slightly flattened sphere to a pancake-like shape ($0.6 < R_0/h < 3$, *h* is the channel height).



FIG. 2. Snapshots of an anchored bubble pulsating under high amplitude of ultrasound (2 μ s exposure, f = 104 kHz). The pit appears as a white disk behind the bubble. During its growth, the bubble explores different shape modes directly related to its radius. The number of peaks corresponds to the most amplified mode for a given bubble radius. (Multimedia view) [URL: http://dx.doi.org/10.1063/1.4942917.1]

Once released into the main channel, bubbles are advected by the surrounding liquid. If the drag force is low enough, capillary forces trap the bubble flowing over the pit. As soon as a bubble gets trapped, the nitrogen pressure is lowered in order to stop the bubble production. Ultrasound emission is then switched on at a frequency that is close enough to the bubble resonance (f = 20-150 kHz) but also matches one of the resonances of the glass bar actuated by the piezo-plate. We then observe the oscillations of the bubble with an inverted microscope (Olympus, model IX70) and record them using a fast camera (Miro 4, Vision Research).

Interestingly, we observed that, even without ultrasound, the bubble radius grows during the time of the experiment, since the liquid contains dissolved nitrogen and is always refreshed, thanks to the liquid flow rate. The growth rate of the bubble radius is typically of $0.1-1 \ \mu m/s$. This allows us to explore the bubble vibrations as a function of its mean radius R_0 under constant acoustic conditions.

As the maximal sampling rate of the camera is much smaller than the driving frequency f of the bubble (f = 20-150 kHz), a 2 μ s exposure time was used to resolve the bubble dynamics. The piezoelectric voltage source was operated at a frequency slightly higher than f to trigger the image acquisition in a stroboscopic way and slow down the bubble dynamics down to 5 Hz ($\Delta f = f - Nf_s = 5$ Hz, with N an integer). This gives access to the high frequency dynamics of the bubble oscillations provided these are periodic, as was separately verified.

C. Image analysis

Utilizing the previous procedure, we can record the overall size evolution and vibration dynamics of a bubble over a long time duration (a few tens of seconds), as illustrated in Fig. 2 (Multimedia view) and also access its short time dynamics.

To analyze the shape of the bubble, a threshold has been first applied on the images to identify its contour. The angular dependency of the radius is extracted for each time step and is decomposed in Fourier modes,

$$\rho(\theta, t) = R_0 + a_0(t) + \sum_{n \ge 1} a_n(t) \cos(n\theta + \psi_n(t)),$$
(1)

where R_0 is the sliding average of the bubble radius over a pulsation and can slowly evolve with the time, $a_n(t)$ is the amplitude of the mode n, and $\psi_n(t)$ the angular orientation phase of the mode, taking the x-axis as reference for the angles. In practice, $|a_n|$ and ψ_n are determined for each image from the discrete Fourier transform of $\rho(\theta, t)$ made on the angle θ . Then the rotation of the mode is extracted from $\psi_n(t)$ by considering the angle of the first peak of the mode n encountered counterclockwise: $\theta_n = -\psi_n/n$. The amplitude $a_n(t)$ of the mode can finally be recovered. Its Fourier transform leads to a full description of the bubble shape, and particularly to the fundamental frequency of the mode ω_n and its harmonics. Whenever harmonics can be neglected, we simplify the expression of $a_n(t)$ in

$$a_n(t) = A_n \cos(\omega_n t + \phi_n), \tag{2}$$

where A_n is the absolute amplitude of the mode n, and ϕ_n its temporal phase. Note that A_n and ϕ_n may slowly vary during the bubble growth process or a transient regime of the pulsation. For this reason, we used synchronous demodulation by multiplying $a_n(t)$ by $\cos(\omega_n t)$ in order to get the time evolution of A_n and ϕ_n .



FIG. 3. Breathing mode: Rescaled amplitude A_0/A_0^{max} (a) and temporal phase shift $\phi_0 - \phi_{ref}$ (b) of the mode 0 for three different frequencies: f = 30, 40, and 50 kHz. For each frequency, a different range of bubble radii has been explored. A master curve is obtained by using $R_0 f$ as x-axis, with R_0 the bubble mean radius and f the driving frequency. The elastic wall 2D model fit (dashed line) gives access to the velocity of the Rayleigh waves at the water/PDMS interface: $c_R = 40$ m/s.

III. AXISYMMETRIC PULSATION AT LOW EXCITATION

A. Results on bubble resonance

For a given driving frequency f, the vibration of a single bubble anchored on its pit has been analyzed: at low amplitude of excitation, meaning for a driving pressure less than a few 10 kPa, only the breathing mode n = 0 gets excited.

Using successive sinusoidal burst signals of duration $T_{burst} = 850$ ms with a time interval of 1 s generated by the piezo-plate, we measured the amplitude A_0 and the temporal phase ϕ_0 for different values of the bubble radius. We repeated this experiment for different frequencies (f = 30, 40, and 50 kHz), without changing the value of the piezo-voltage (A = 10 V). We plotted the results as a function of the product $R_0 f$ in order to get the master curve shown on Fig. 3. To get rid of the phase shift induced by the piezo, we chose for each frequency a reference phase ϕ_{ref} to get a match of the three curves.

We find that the resonance of the cylindrical bubble is obtained for $R_0 f = 1.5$ m/s, which is half of the Minnaert resonance given by a spherical bubble model. Moreover, the phase shift between low and high $R_0 f$ stays around $\pi/2$, instead of the π shift obtained in the 3D model.¹³

B. Model for the 2D pulsation of a bubble

The question of the bubble 2D pulsation has been considered theoretically by Prosperetti¹⁴ for a bubble surrounded by an incompressible liquid. In the case of an infinite liquid channel, because of the divergence of the inertial mass of liquid around the bubble, this author used a cutoff parameter *S* for the liquid mass radius. Physically, it corresponds to the situation of a cylindrical bubble surrounded by a shell of incompressible liquid with a diameter *S*. In that case, considering small oscillations around the equilibrium radius $R(t) = R_0(1 + X(t))$, with $X(t) = a_0(t)/R_0 \ll 1$, the linearized 2D Rayleigh-Plesset equation is written,

$$\ln\left(\frac{S}{R_0}\right)\ddot{X} + \frac{2\kappa p_0}{\rho_f R_0^2}X = -\frac{P_{ac}}{\rho_f R_0^2},$$
(3)

where ρ_f is the density of the fluid, p_0 the inner gas pressure of the bubble, κ its polytropic index, and P_{ac} the driving acoustic pressure.

The cylindrical bubble is also subject to damping. For a spherical bubble with a radius between 15 and 75 μ m, the radiation and thermal damping terms dominate the viscous damping.³ But in a PDMS microchannel, another radiation damping term can also arise when considering the elasticity of the walls and the surface wave propagation at the water/PDMS interface generated by the bubble pulsation. This mechanism sketched in Fig. 4 was already suggested in Ref. 10. In Sec. III C, we will include the wall deformation to the classical 2D model in order to quantify this 2D radiation damping and understand the origin of the resonance shift.



FIG. 4. (a) Schematic representation of the bubble pulsation: the pressure variations in the liquid induce a deformation of the elastic PDMS walls, the deformation being accentuated here for a better understanding, and generate Rayleigh waves at the water/PDMS interface with a wavelength $\lambda_{Rayleigh}$ of a few hundred micrometers. (b) Schematic representation of a shape mode n = 4. The solid and dashed lines correspond to the extrema of vibration of the mode. $\rho(\theta, t)$ is the distance between the center of the bubble and the periphery of the bubble at a time t. The amplitude of vibration of the liquid away from the bubble (blue arrows) is vanishing in the angular direction of the nodes (thin dashes), and is maximum at the antinodes.

C. Model for the 2D pulsation of a bubble confined between elastic walls

For simplification, we first assume that the liquid pressure p creates a small deflection on the elastic walls of the channel,

$$h = h_0 + \alpha \frac{p}{E} h_0, \tag{4}$$

where h_0 is the thickness of the channel at rest, E the Young's modulus of the PDMS, and α a geometric parameter.

In the following, we also suppose that the bubble wall pushes the liquid radially outward with no significant variation in the height of the channel. Thus, the velocity of the liquid around the bubble is close to a parallel plug flow over the channel thickness, meaning that its radial and tangential components u_r and u_θ do not depend on the vertical position z in the channel. Including the vertical wall deformation, the conservation of mass links the liquid velocity (u_r, u_θ) and the thickness h of the channel as follows:

$$\frac{\partial h}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}(rhu_r) + \frac{1}{r}\frac{\partial}{\partial \theta}(hu_\theta) = 0.$$
(5)

In addition, the conservation of momentum for small amplitudes of motion leads to an Euler equation, which is written in polar coordinates,

$$\rho_f \frac{\partial u_r}{\partial t} = -\frac{\partial p}{\partial r},\tag{6}$$

$$\rho_f \frac{\partial u_\theta}{\partial t} = -\frac{1}{r} \frac{\partial p}{\partial \theta},\tag{7}$$

where *p* represents the pressure in the liquid.

By replacing h from Eq. (4) and keeping only first order linear terms in Eqs. (5)-(7), we obtain a d'Alembert wave equation

$$\frac{1}{c_{\mathbf{p}}^{2}}\frac{\partial^{2}p}{\partial t^{2}} - \frac{\partial^{2}p}{\partial r^{2}} - \frac{1}{r}\frac{\partial p}{\partial r} - \frac{1}{r^{2}}\frac{\partial^{2}p}{\partial \theta^{2}} = 0,$$
(8)

where $c_{\mathbf{R}} = (E/\alpha \rho_f)^{1/2}$ is the velocity of the surface waves generated by the liquid pressure in the channel. These waves are usually called Rayleigh waves and their velocity is written: $c_{\mathbf{Rayleigh}} \approx \gamma \sqrt{G/\rho_f}$, where G is the shear modulus of the material with a prefactor γ in the range 0.87-0.95, depending on Poisson's ratio of the material.¹⁵ In the case of a PDMS wall, E = 3Gleading to $\alpha \approx 3$. To summarize, the bubble pulsation induces an oscillatory variation of the liquid pressure close to the bubble wall. This pressure variation excites Rayleigh waves at the PDMS/water interface by pressing periodically the elastic wall. For a given pulsation ω , we can write $p = \text{Re}(\overline{p}e^{j\omega t})$. As the bubble is alone in the channel and its radius is much smaller than the channel width, we can consider that the driving pressure field is uniform, and because of the axisymmetry of the breathing mode, that \overline{p} does not depend on θ . Then Eq. (8) becomes a Helmholtz equation in cylindrical coordinates,

$$k^{2}\overline{p} + \frac{\partial^{2}\overline{p}}{\partial r^{2}} + \frac{1}{r}\frac{\partial\overline{p}}{\partial r} = 0,$$
(9)

where $k = 2\pi/\lambda = \omega/c_{\mathbf{R}}$. The solution for an outgoing propagating wave has the following form:¹⁶

$$\overline{p} = \beta_0 H_0^{(2)}(kr),$$

where $H_0^{(2)} = J_0 - jN_0$ is the Bessel function of the third kind, also known as the Hankel function, and β_0 a constant to determine. Superscript (2) of the Hankel function refers to the outwardpropagating wave. It is constructed from the Bessel functions of the first kind J_0 , and of the second kind N_0 .

The expression of the radial fluid velocity $u_r = \operatorname{Re}(\overline{u}_r e^{j\omega t})$ can be obtained from Eq. (6),

$$\overline{u}_r = \frac{-1}{j\omega\rho_f} \frac{\partial\overline{p}}{\partial r} = \frac{j\beta_0 k}{\rho_f \omega} H_0^{\prime(2)}(kr).$$
(10)

By developing Eq. (10) at $r = R_0$ and writing the bubble oscillation $X = \text{Re}(\overline{X}e^{j\omega t})$, we can deduce the expression of β_0 and get

$$\overline{p} = \frac{\omega^2 \rho_f}{k} \frac{H_0^{(2)}(kr)}{H_0^{\prime(2)}(kR_0)} R_0 \overline{X}$$

The liquid pressure near the bubble is

$$p_L = p_0 + \overline{p} e^{j\omega t} + \overline{P_{ac}} e^{j\omega t}.$$
(11)

The pressure is also linked to the pressure p_g in the bubble with a correction due to capillarity (Laplace pressure) and a second correction due to viscosity,

$$p_L = p_g - p_{cap} - p_{vis}.$$
 (12)

The gas pressure in the bubble is given by a polytropic law $p_g = p_{g,0}(R/R_0)^{-2\kappa} \simeq p_{g,0}(1 - 2\kappa X)$, where κ is the polytropic index, around 1.4 for air.

Note that we have here neglected the influence of the micropit. In fact, the bubble pulsation does not look affected by the pit as long as the bubble radius is larger than the size of the pit: $R_0 > (d + h)/2$ and as long as a wetting film exists between the bubble and the PDMS wall.

As we are interested in bubbles with a radius larger than 15 μ m and acoustically driven at frequencies smaller than 150 kHz, we are working in a domain where $\sigma/R_0 \ll p_{g,0}$ and $\eta \omega \ll p_{g,0}$. Therefore, we can neglect the capillary and viscous contributions in the equation of the breathing mode.

We finally obtain the pulsation equation for a single bubble by equating two expressions (11) and (12),

$$\left[\rho_f c_{\mathbf{R}}^2 \, k R_0 \frac{H_0^{(2)}(k R_0)}{H_0^{(2)}(k R_0)} + 2\kappa p_0\right] \overline{X} = -\overline{P_{ac}}.$$
(13)

According to Eq. (13), the pulsation of the bubble depends principally on the product kR_0 , which is equivalent to R_0f if the velocity of Rayleigh waves in the PDMS does not vary much within our frequency range. In Fig. 3, all the data have been plotted using R_0f as x-axis. By fitting the experimental data with the elastic wall 2D model presented in Eq. (13), a good match is obtained for the three different frequencies, when the value of the Rayleigh wave velocity $c_{\mathbf{R}}$ is 40 m/s.

Since the typical value of *E* is 0.8–4 MPa for the PDMS, depending on the age and curing time of the elastomer,¹⁷ the value of the fitting parameter $c_{\mathbf{R}}$ leads to an estimate of the geometric parameter $\alpha \in [0.5:2.5]$, which is not that far from the expected value ($\alpha \simeq 3$).

In our experiment, bubbles are small compared to the wavelength, which means $kR_0 = 2\pi R_0/\lambda \ll 1$. In that case, Eq. (13) can be written,¹⁸

$$\left[-\omega^2 \ln\left(\frac{\lambda/2\pi}{R_0}\right) + \frac{2\kappa p_0}{\rho_f R_0^2} + j\frac{\pi}{2}\omega^2\right]\overline{X} = -\frac{\overline{P_{ac}}}{\rho_f R_0^2}.$$
(14)

The natural pulsation of this equation can be compared to the one of the classical 2D models (Eq. (3)). For small radii, we have the remarkable result that the cutoff length in the classical 2D model is given by the wavelength λ of the Rayleigh wave,

$$S = \lambda/2\pi$$

The bubble behaves as if it was confined in an annulus of liquid whose size scales like the Rayleigh wave in the PDMS.

Note that Eq. (14) also includes an additional term in the square brackets compared to the classical 2D model (Eq. (3)). This term corresponds to the 2D radiation damping due to the generation of Rayleigh waves. Because of this radiation damping, the phase tends to $\pi/2$ instead of 0 as $R_0 f$ tends to infinity, so that the total phase shift between low and high values of $R_0 f$ is $\pi/2$. This is consistent with the data shown in Fig. 3(b).

To summarize, even for small bubble radii, the Rayleigh wave has to be included in the model. It modifies both the effective mass of the bubble by adding some confinement around the bubble, and the radiation damping term. This affects principally the resonant radius of the bubble and the temporal phase shift of the mode 0 above the resonant radius. The effective quality factor of the bubble resonator finally depends on the parameter kR_0 : the higher the Rayleigh wave velocity, the higher the quality factor.

IV. PARAMETRIC INSTABILITY AT HIGH EXCITATION

A. Amplitude and rotation of the bubble shape modes

For higher amplitudes of excitation, the bubble exhibits different deformation modes, indicated by the number *n* of peaks around its periphery. The shapes shown on Fig. 2 (Multimedia view) are recorded during the growth process of a same anchored bubble, for a total duration of 60 s. We could generally observe modes going from n = 2 to n = 12.

We analyzed the vibration of each mode. Fig. 5 is an illustration of the time evolution of the modes 0 and 4 of a bubble with a radius $R_0 = 24 \ \mu m$, shortly after switching on the sound excitation. We first observe the response of a mode 0 characterized by an axisymmetric oscillation of the bubble and, after several tens of excitation periods, the appearance of the most amplified mode n = 4. Note that the appearance of this mode 4 takes also many periods.

Moreover, whereas the amplitude of the mode 0 oscillates at the same frequency as the driving frequency f, the amplitude of the mode n oscillates at a frequency f/2. Indeed, we see in Fig. 5 that the period of mode 4 is twice that of mode 0. More generally, we observe this period doubling whenever a mode n becomes dominant compared to the other modes n > 0. This is the characteristic of a parametric instability.



FIG. 5. Time evolution of the breathing mode (top curve) and the most amplified mode n = 4 (bottom curve) of a pulsating bubble with a given radius $R_0 = 24 \ \mu m$ and excited at a frequency $f = 113 \ \text{kHz}$. Time is non-dimensionalized with the stroboscopic frequency.



FIG. 6. Vibration modes of a growing bubble excited at a fixed frequency f = 104 kHz with a fixed amplitude A = 22.5 V corresponding to a driving pressure $P_{ac} \approx 45$ kPa: (a) Maximal amplitude of the breathing mode a_0^{max} (black line) and absolute amplitude of each mode $|a_n(t)|$ as a function of the time, for an increasing radius R_0 . (b) Orientation $\theta_n/(2\pi)$ (revolutions) of the dominant mode n at a given bubble radius R_0 . A rotation of the mode sometimes appears during the transition between the modes but also when the mode is stable. The rotation speed is of order 0.5 revolution/s. The grey zones correspond to uncertainty domains due to the appearance of a new peak during the transition; inset: time evolution of the bubble mean radius R_0 .

For longer time scales, Fig. 6 illustrates the results of the same analysis performed on a steadily growing bubble, covering a continuous variation of radii. For a constant driving frequency of 104 kHz and a given amplitude of excitation corresponding to a piezo-excitation of 22.5 V, Figs. 6(a) and 6(b) show the time evolution of the absolute amplitude and of the orientation of the dominant modes while the bubble radius grows (see inset in Fig. 6(b)). As we are above the resonance frequency, the amplitude of the breathing mode decreases with the increasing radius, and thus with the time (see the black line on Fig. 6(a)), as predicted by our breathing mode model. The absolute amplitude $|a_n|$ of the dominant modes is obtained using Eq. (1) and is also represented in Fig. 6(a). It shows that the bubble selects a preferred mode, depending on its radius. When the radius increases, the bubble accommodates the same mode until a transition occurs toward the next mode. With the time resolution, we could not see any coexistence of the modes, apart from the breathing mode. However, the transition from a mode to another one seems to slightly change the amplitude of the mode 0. Moreover, we notice that the amplitude of each mode decreases with time.

The angular orientation of the most amplified mode, given by $\theta_n = -\psi_n/n$, is reported in Fig. 6(b). It represents the angle of the first peak of the mode encountered counterclockwise. Thus, for each mode, one can reconstruct the angle of each peak by adding k/n, with k < n (k an integer), to the angle plotted in Fig. 6(b). The angle of the mode has been unwrapped in order to better see the rotation. During the transition between two following modes, a new peak appears on the periphery of the bubble. However, the time resolution does not allow us to determine the precise location of the appearance of this new peak or observe the rearrangement of the preexisting peaks. Therefore, grey zones have been added on the figure to take into account this reorientation of the mode due to this new peak. The amplitude of these grey zones is thus given by 1/n. One could think that the presence of the pit close to the bubble wall influences the orientation of the surface modes. But the angle θ_n evolves continuously in Fig. 6(b), meaning that the surface mode is free to rotate, even when the bubble wall is close to the pit. Most of the time, the rotation occurs during the transition between two modes. But sometimes, it happens far away from the transition. The angular rotation speed is typically 0.5 revolution/s and seems to be constant for all the modes. Both directions of the rotations (clockwise and counterclockwise) have been observed.

By controlling the amplitude of the driving acoustic pressure, we were able to build a phase diagram of the different shapes encountered by a quasi-2D bubble, see the symbols in Fig. 7. For all amplitudes, the bubble oscillates with an axisymmetric mode (n = 0). By fitting the radius evolution of a_0 with Eq. (13), we have access to the Rayleigh wave velocity in the device $c_{\mathbf{R}} = 44$ m/s, and also to an estimate of the conversion factor between the voltage A applied to the piezo and the acoustic driving pressure P_{ac} exciting the bubble: P_{ac} (kPa) $\approx 2 A$ (V). In addition to this breathing



FIG. 7. Experimental phase diagram showing the most amplified mode as a function of the bubble radius R_0 and the driving pressure P_{ac} (kPa). Experimental data are reported by black crosses whenever the mode 0 is the only mode excited. In the other cases, the breathing mode is superimposed with a unique surface mode: n = 4 (cyan squares), n = 5 (light blue triangles), n = 6 (dark blue circles), n = 7 (purple downward-pointing triangles), or n = 8 (mauve diamonds). The colored dashed lines correspond to the theoretical resonant radius of each surface mode, obtained with Eq. (19) for a surface tension $\sigma = 25$ mN/m. Colored domains correspond to the theoretical domains of each surface mode, the mode number being written in each domain. They are obtained with Eqs. (22) and (23), with a Rayleigh wave velocity $c_{\mathbf{R}} = 44$ m/s, a surface tension $\sigma = 29$ mN/m and a damping constant $\gamma = 1.9 \times 10^5$ s⁻¹. The black domain corresponds to a zone where only the mode 0 is excited. In the colored areas, both the breathing mode and a unique shape mode are excited.

mode, starting above a certain acoustic threshold, the bubble undergoes the shape instability. The amplitude of the threshold varies with the bubble radius, showing a minimum for the mode n = 5, at a radius clearly above the theoretical resonance radius of the breathing mode $R_0^{res} = 15 \ \mu m$.

B. Model for the natural pulsation of the modes

For small amplitudes of oscillations ($a_0 \ll R_0$), we now perform the same analysis as for the breathing mode, but this time to describe surface modes. Considering only a nonrotating amplified mode *n*, the bubble radius is written as $\rho(\theta, t) = R(t) + a_n \cos(n\theta)$, with $R(t) = R_0 + a_0(t)$, according to Eq. (1). The researched solution \overline{p} now depends on θ . The solution of Eq. (8) for an outgoing propagating wave has the form,

$$\overline{p} = \beta_n H_n^{(2)}(kr) \cos(n\theta), \tag{15}$$

where $H_n^{(2)}$ is the Hankel function of order n^{19} and β_n a constant to determine. From the Euler equation (Eq. (6)), we obtain the radial fluid velocity,

$$\overline{u}_r = \frac{-1}{j\omega\rho_f}\frac{\partial\overline{p}}{\partial r} = \frac{j}{\rho_f\omega}\beta_n k H_n^{\prime(2)}\cos(n\theta).$$

We notice that the velocity of the liquid vanishes when the angular direction is that of the surface nodes (see Figure 4(b)).

At the bubble surface, this velocity should be equal to the bubble wall velocity $u_r = \dot{R} + \dot{a}_n \cos(n\theta)$: this enables to find the constant β_n . The total liquid pressure (see Eq. (11)) expressed at the bubble surface is thus written,

$$p_L = p_0 + \frac{\rho_f \omega^2}{k} \frac{H_n^{(2)}(kR_0)}{H_n^{(2)}(kR_0)} \overline{a}_n \cos(n\theta) e^{j\omega t} + P_{\rm ac}(t).$$
(16)

It is also linked to the gas pressure by the Laplace equation with an interface curvature that is the sum of the polar curvature $\kappa_{polar} = |\rho^2 + 2(\partial_\theta \rho)^2 - \rho \partial_\theta^2 \rho|/(\rho^2 + (\partial_\theta \rho)^2)^{3/2}$ in the plane, and

of the transverse curvature in the thickness, $\kappa_{trans} = 2/h$. Thus, we can also write the total liquid pressure as

$$p_L = p_g - \frac{\sigma}{R_0} - (n-1)(n+1)\frac{\sigma}{R_0^2}a_n\cos(n\theta) - \frac{2\sigma}{h} + p_{n,vis}.$$
 (17)

Note that surface tension terms are not negligible when considering the modes. In the following, we drop the viscous terms for simplicity.

We obtain the dynamics of the modes from Eqs. (16) and (17),

$$\left[\frac{\rho_f \omega^2}{k} \frac{H_n^{(2)}(kR_0)}{H_n^{\prime(2)}(kR_0)} + (n-1)(n+1)\frac{\sigma}{R_0^2}\right]\overline{a}_n = -p_n^{\text{ext}},$$

where p_n^{ext} is linked to the non-uniformity of the acoustic field, here assumed to be 0.

The natural frequency of the mode is thus

$$\omega_n^2 = \frac{-kR_0}{\operatorname{Re}\left(\frac{H_n^{(2)}(kR_0)}{H_n^{(2)}(kR_0)}\right)} \quad \frac{(n-1)(n+1)\sigma}{\rho_f R_0^3}.$$
(18)

In our experiments, the maximum value of kR_0 is 0.8. For small bubbles with respect to the wavelength ($kR_0 \ll 1$), the first factor on the right hand side of Eq. (18) tends²⁰ to *n* and consequently the natural pulsation tends to

$$\omega_{n}^{2} = n(n-1)(n+1)\frac{\sigma}{\rho_{f}R_{0}^{3}},$$
(19)

which is the natural pulsation of the mode n in the classical 2D theory.²¹

To summarize, the natural pulsation of the shape modes has no specific dependence on the channel-wall elasticity. In the opposite, the breathing mode always depends on the wavelength of the Rayleigh waves, even for small values of the bubble radius. Assuming that there is no damping, the resonance of the parametric mode will occur when $R_n^{res} = \left(n(n-1)(n+1)\frac{\sigma}{\rho_f(\omega/2)^2}\right)^{1/3}$, since the driving pulsation ω is fixed and equals $2\omega_n$. The theoretical resonant radius R_n^{res} of each surface mode has been indicated in Fig. 7, for a surface tension $\sigma = 25$ mN/m. They show a good agreement with the experimental resonant radii obtained at all driving pressure.

C. Model for large amplitude oscillations

At *large amplitudes of oscillations*, the analysis becomes very complex if we keep considering the channel-wall elasticity. We will thus remain in the case $kR_0 \ll 1$ and assume that the channel walls are rigid. In this case, the oscillation flow field around the bubble in 2D can be described by the potential field,

$$\varphi = \dot{R}R\ln(r/R_0) - \sum_{n\geq 1} \frac{b_n}{r^{n+1}}\cos(n\theta),$$

where b_n is a dimensional coefficient to determine. The velocity field $\mathbf{v} = \nabla \varphi$ has a radial component,

$$u_r = \frac{RR}{r} + (n+1)\sum_{n\geq 1}\frac{b_n}{r^{n+2}}\cos(n\theta).$$

From the boundary conditions (Eq. (1)), we obtain the coefficients b_n as a function of a_n : $b_n = \dot{a}_n R^{n+2}/(n+1)$. Neglecting dissipation for simplicity, the dynamics is obtained by writing the Bernoulli equation that provides the pressure,

$$p_L = p_0 + P_{ac}(t) + \rho_f \left[\frac{\partial \varphi}{\partial t} + \frac{1}{2} (\nabla \varphi)^2 \right]_R^S,$$
(20)

with $S \gg R_0$.

We use as before the fact that pressure is given at the interface by the Laplace law in Eq. (17), which closes the problem. Writing the dynamics of each bubble mode, we arrive in the case of small shape oscillations $(a_n/R_0 \ll 1)$ at

$$\ddot{a}_n + 2\frac{\dot{R}}{R}\dot{a}_n + \left[\omega_n^2 - (n-1)\frac{\ddot{R}}{R}\right]a_n = 0.$$
(21)

Following Ref. 22, we introduce a new variable $c_n = a_n R/R_0$. For a sinusoidal pulsation of the radius, $R = R_0 + A_0 \cos(\omega t)$, we get to the first order,

$$\ddot{c}_n + \omega_n^2 \left[1 + n \frac{A_0}{R_0} \left(\frac{\omega}{\omega_n} \right)^2 \cos(\omega t) \right] c_n = 0,$$
(22)

which is a Mathieu equation. This kind of equation is known to have, amongst others, unstable solutions with period-doubling,²³ the varying parameters being in the case of Eq. (21): ω_n and $h = n \frac{A_0}{R_0} (\frac{\omega}{\omega_n})^2$. These solutions become unstable above a threshold given by $h_t = 2 \left| 1 - \left(\frac{\omega}{2\omega_n} \right)^2 \right|$. Thus, one needs to add damping terms in Eq. (22) in order to get a nonzero threshold at $\omega = 2\omega_n$, as observed experimentally. If we add a damping term $\gamma \dot{c}_n$ in Eq. (22), the instability threshold then becomes

$$h_t = 2\sqrt{\left[1 - \left(\frac{\omega}{2\omega_n}\right)^2\right]^2 + \frac{\omega^2 \gamma^2}{4\omega_n^4}}.$$
(23)

Using Eqs. (13) and (19) to link the two parameters ω_n and *h* to the experimental parameters R_0 and P_{ac} , the theoretical existence domains of each mode have been plotted in Fig. 7, for a damping constant $\gamma = 1.9 \times 10^5 \text{ s}^{-1}$ and a surface tension $\sigma = 29 \text{ mN/m}$. We obtain a pretty good agreement between these domains and the experimental data. We recover a "Mathieu's tongue" for each surface mode, the asymmetry of the tongues coming from the fact that ω_n is proportional to $R_0^{-3/2}$. The theoretical pressure threshold of the mode n = 4 is lower than expected, meaning that the damping constant γ should depend on the bubble radius R_0 and the surface mode n.

V. CONCLUSIONS

In this paper, we proposed an experimental and theoretical study of the acoustic response of a flattened bubble at rest in a microfluidic channel. Such a bubble pinned on a micropit presents axisymmetric oscillations for reduced sound amplitudes, and a combination of axisymmetric and shape oscillations above some critical threshold of the exciting field. To understand this, we derived a 2D modified Rayleigh-Plesset equation taking into account the elasticity of the channel walls. It is found that the presence of the two confining walls leads principally to a modification of the breathing mode. More precisely, we show that the radiation damping due to the Rayleigh wave emission at the water/PDMS interface is responsible for the diminution of the bubble resonance frequency.

As long as the bubble radius is small compared to the Rayleigh wavelength, the wall deformation does not affect much the shape modes dynamics, and the general form of the 2D Rayleigh-Plesset equation is sufficient to understand it. We show that the 2D dynamics of the parametric shape modes is governed by a Mathieu equation in which a damping term was added to account for the nonzero driving pressure threshold of the modes. The magnitude of this damping term is too high to correspond to a 2D radiation damping. To seek for the origin of this damping, a calculation of the 2D viscous damping could be the object of future studies. Note that Leidenfrost droplets confined in between two planes also exhibit spontaneous 2D modes,²⁴ but without any parametric forcing, suggesting a different mechanism.

In order to give a physical picture of the involved dynamics under 2D confinement, we could say that the bubble oscillation is partly restricted by the confinement which limits its quality factor to low values. This may be due to the fact that, though a wetting film separates the bubble from the walls, the bubble oscillations are limited not only by the liquid inertia in 2D (see Ref. 14) but also

by some surface wave emission. Thus, from an energy perspective, one may expect that the energy dissipation does not only occur via the liquid friction or the energy transfer between the breathing and the surface mode (see Refs. 25 and 26 for a discussion). It also occurs by some surface wave emission.

In conclusion, we think that we have captured the main dynamics of these flattened bubbles and highlighted the role of the elasticity of the walls in which they are confined.

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- ¹⁸ Using asymptotic values of the Hankel functions and its derivative, we can show that $-H_0^{(2)}(x)/(xH_0^{(2)}(x))$ tends to $\ln(x) + j\pi/2$ for vanishing values of $x = kR_0$.
- ¹⁹ For large distance to the bubble, we have $\lim_{k \to \infty} H_n^{(2)}(kr) = \sqrt{\frac{2}{\pi k r}} e^{-j(kr n\pi/2 \pi/4)}$, meaning that the pressure and velocity field decay with the same algebraic exponent whatever the mode number $(n \ge 0)$. ²⁰ Here again we use the asymptotic values of the Hankel functions: $-H_n^{(2)}(x)/(xH_n'^{(2)}(x))$ tends to 1/n for vanishing
- values of x, in the case $n \ge 1$.
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