

## Observation of bi-stable and chaotic bubble dynamics

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### Introduction

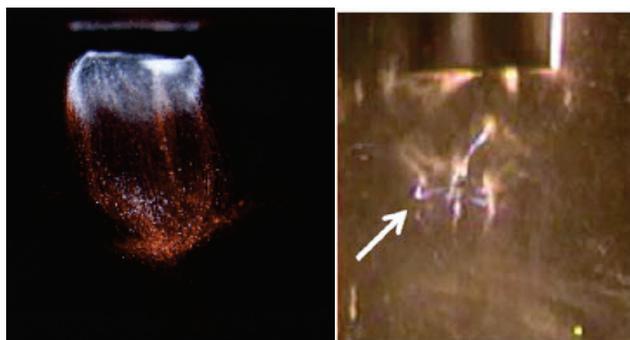
The dynamics of an acoustically driven spherical bubble can be involved due to the nonlinear equations of motion [1]. Consequences of nonlinearity comprise non-harmonic volume oscillations and the strong bubble collapse that can cause extreme compression of the gas phase leading to heating, chemical reactions, and sonoluminescence (SL) [2]. Further phenomena that are predicted by analysis and numerical calculations are nonlinear resonances [3,4], different bifurcations, multi-stability, and deterministic chaos [5]. Although some experiments in bubble traps have documented the existence of period doubling and aperiodic oscillations [6], the direct preparation and observation of such dynamical details with “levitated bubbles” remains difficult, mainly because of stability and parameter restrictions. Thus the question persists if the nonlinear features are “real” and how well they are captured by simple spherical bubble models.

Here we report on recordings of individual bubbles in multi-bubble fields driven around 23 kHz in acids that exhibit bi-stability, i.e., a hysteretic behavior of large and small volume oscillation amplitude. The hysteresis is coupled to distinct spatial motion and partly to SL emissions. In one observed case (A) in sulfuric acid [7,8], the small oscillation state interrupts the fast bubble translation of the large expansion state. While the smaller pulsations show additional surface mode oscillations, the larger pulsations are spherical apart from strong jetting during collapse. The stagnant, smaller amplitude oscillation state ends after a while through an increase of bubble expansion, and the large amplitude and fast moving dynamics resumes. The second case (B) is observed in phosphoric acid, and the bubble motion in space is reciprocating (alternately several oscillations forth and then several periods back) [9]. The larger oscillation state is again spherical with strong collapse jets, but the smaller oscillation state appears aperiodic or chaotic. The findings are partly reproduced by numerical model calculations with a Keller-Miksis model that couples to a one-dimensional spatial translation. For capturing of case B, it appears necessary to include the feedback pressure coupling of translation on the bubble driving pressure [10].

### Experiments

The experimental setup consists of an inhouse made 1 cm diameter titanium sonotrode that dips from top into a cubic glass cell (5 cm side length). The cell is filled with about 100 ml of concentrated H<sub>2</sub>SO<sub>4</sub> (case A) or H<sub>3</sub>PO<sub>4</sub> (85%, case B)

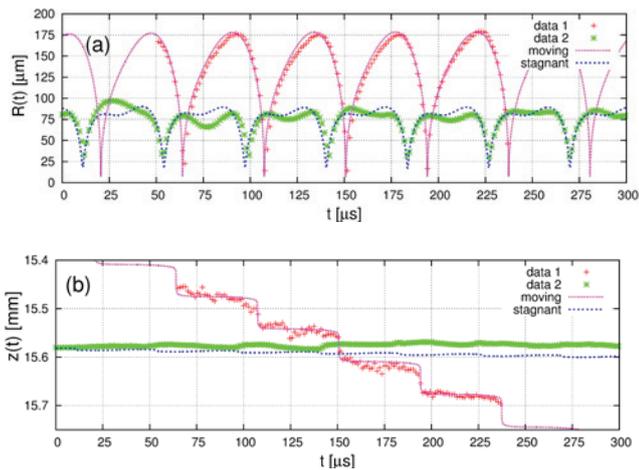
and covered with a drilled Teflon cap. The liquids are saturated with xenon by bubbling the gas through, and in case A the salt NaSO<sub>4</sub> is additionally dissolved in the sulfuric acid (0.1 M). The sonotrode operates around its main resonance of 23 kHz and is driven sinusoidally by a function generator via a power amplifier. At higher driving voltages, cavitation with intense SL occurs in both cases, visible at normal room light and colored in case A (Fig. 1). Bubble dynamics are recorded by a high-speed video camera (Photron APX-RS) with background illumination (inhouse made LED lamp).



**Figure 1:** Left: Digital photograph of colored SL in case A. Right: Video frame of intense SL in case B. The arrow indicates the approximate position of the reciprocating bubbles. The sonotrode tip is visible on top.

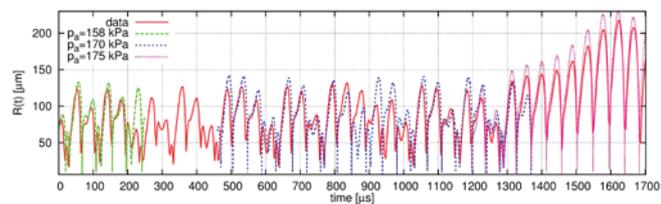
### Bubble dynamics

A comparison of observed high-speed data with model simulations after parameter fit is shown in Fig. 2 for case A. The large amplitude oscillation (red crosses for measurement and pink dotted line for simulation) is accompanied by fast forward jumps and jetting. Although the jetting is not included in the purely spherical bubble model, the numerical simulation can well reproduce oscillation and translation for the fitted parameters (equilibrium bubble radius  $R_0 = 60 \mu\text{m}$ , piston source sound field with local pressure amplitude at bubble position  $p_a = 95 \text{ kPa}$ ). This is also true for the second, coexisting type of bubble dynamics, shown by green stars (experiment) and blue dashed lines (numeric). Here the collapse is less violently, and the observed bubble shows surface oscillations (unstable modes) in superposition of the volume oscillations. Surface modes are not captured in the model, but again the main oscillation and the nearly stagnant translation are well reproduced.



**Figure 2:** Radius-time dynamics (a) and spatial position  $z$  vs. time (b) of both coexisting bubble oscillation forms, as seen in the experiment case A (crosses and stars, “data”) and in the numerical model (dotted and dashed lines) for  $R_0 = 60 \mu\text{m}$  and  $p_a = 95 \text{ kPa}$ .

The second case (B) is presented in Fig. 3. Here a bubble is reciprocating within a certain spatial region, apparently passing varying levels of excitation pressure amplitude. The shown sequence covers the end of the “upward” motion (towards the sonotrode tip) when the radial oscillation suddenly expands successively over several periods. At this moment, a “downwards” motion (away from the sonotrode tip) sets in which contains similar collapse jumps and jetting as seen in case A.



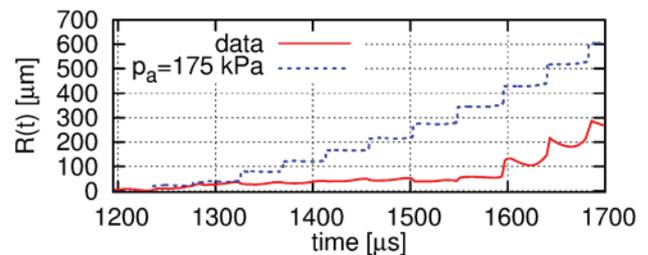
**Figure 3:** Radius-time dynamics of a bi-stable bubble as observed in the experiment case B (red line). Numerical fits have been obtained for  $R_0 = 36.5 \mu\text{m}$  and  $p_a$  in the range from 158 kPa to 175 kPa (green, blue and pink lines).

A numerical reproduction of the observed radius-time data succeeds surprisingly well, including higher- or aperiodic sequences, if the equilibrium radius and the driving pressure are chosen suitably. The translational dynamics, however, is not captured that well by the model. For all pressure values that lead to a good fit, the bubble motion in the model is “downwards”, not only for the highest pressure (for times after about  $t = 1300 \mu\text{s}$ ). The rise of amplitude due to translation towards the sonotrode is therefore mimicked in the simulation by momentarily fixed amplitudes of 158, 170 and 175 kPa along the experimental sequence (Fig. 3). Doing so, the radial dynamics is reasonably well reproduced (apart from a small scale factor).

A closer inspection of the case B simulation shows that the experimentally observed coexistence of the chaotic (nearly period-5) oscillation behavior and the expanded period-1

oscillation is not seen in the model if the coupling term of translational velocity to driving pressure amplitude is neglected), even for an extended variation of initial conditions. This is different in case A, where a coexistence of both solutions persists for uncoupled oscillation and translation. Thus the coupling term appears to be essential for an explanation of the observations.

With respect to the spatial translation in case B, it is shown in Fig. 4 that the details of the simulation at 175 kPa are still not in excellent agreement with the recordings, although the direction of motion comes out right. While the numerics predicts plateaus in-between the jumps, the experimental data show a pronounced back translation (loops). This might be due to the assumption that the sound field corresponds to the analytical solution of a circular piston source on its symmetry axis. Since the reciprocating bubble was actually located more sideways from the symmetry axis, the phase relation between acoustic pressure (determining oscillation) and its gradient (responsible for the translation) might be shifted, leading to different forces on the bubble.



**Figure 4:** Spatial distance  $r(t)$  from a reference position vs. time for the reciprocating bubble dynamics near the point of sudden increase of radial oscillation amplitude and accompanying inversion of translation direction (compare Fig. 3). Increasing  $r(t)$  corresponds to “downwards” motion. Experimental data shown by red continuous line, numerical results (bubble position on the symmetry axis of a piston source) shown by blue dashed line.

## Discussion and conclusion

We have reported two cases of hysteretic bi-stable bubble dynamics in a sonotrode system that generates acoustic cavitation in acids. Both cases have been simulated by a numerical model that couples spherical bubble oscillation with spatial translation in a field of a piston source. Case A shows intermittent switching between two dynamical states: small amplitude oscillation with slow translation, and large amplitude oscillation with fast translation. In the model, both states coexist also without inclusion of translational coupling. Case B documents a reciprocating bubble translation connecting smaller and chaotic amplitude oscillation with upward motion, and larger periodic amplitude oscillation with downwards translation. The model can reproduce well the radial bubble pulsations, but predicts partly wrong translational behavior. This might be due to simplifying assumptions on the spatial structure of the driving sound field. Nevertheless, the model gives

indications that the coupling of translation onto the bubble driving pressure plays an essential role for the bi-stability in case B, and thus should be definitely included modeling and analysis of bubble motion and translation in cavitation fields.

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