

Full Length Research Paper

Stability and dancing dynamics of acoustic single bubbles in aqueous surfactant solution

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Acoustic single bubbles in aqueous solutions of sodium dodecyl sulfate (SDS) have been studied. In dilute SDS solutions, the trapped bubbles are found to be stabilized and sustainable up to higher driving acoustic amplitude. Stroboscopic images of the bubble are consistent with theory, indicating dancing motion and intensive near-resonance oscillations of the bubble. A trace analysis of the bubble fluctuations reveals an oscillatory behavior of 'inverted pendulum' around the pressure antinode.

Key words: Acoustic bubbles, sonochemistry, sodium dodecyl sulfate, cavitation.

INTRODUCTION

Acoustically generated cavitation and bubble dynamics are important topics in various fields of physics and chemistry, ranging from the study of propeller erosion to sonochemistry and biomedical applications (Leighton, 1997). Under a stationary sound field applied in liquid, a single bubble can be generated and trapped at a node (or an antinode) of the standing wave of the acoustic pressure. Such a bubble is dynamically oscillating (that is, shrinking and expanding) in time, synchronized to the acoustic wave, but fixed at a specific position and stable over many periods. This stability is especially advantageous and has played an essential role in the detailed optical investigation of processes happening inside collapsing bubbles, such as sonoluminescence (Gaitan, Crum, Church and Roy, 1992; Matula and Roy, 1997).

Since the surface tension and the thermodynamic stability of the gas-liquid interface are critical in bubble dynamics, an addition of a surfactant into the liquid is expected to alter the stability of acoustic bubbles. Surfactant molecules may concentrate and align themselves on the bubble surface to form so-called 'dirty bubbles', so that they not only change the macroscopic interface property but also have profound microscopic effects on intermolecular energy transfer and relaxation processes during the rapid contraction and collapse. For example, dilute organic solutes are found to quench sonoluminescence by offering an energy-dissipating path of chemical

decomposition in the interior vapor (Ashokkumar et al., 2000). Among other known effects of surfactants on bubbles are: stabilization of bubble nuclei, acceleration of bubble growth, suppression of bubble coalescence, and modification of acoustic microstreaming (Crum, 1980; Ashokkumar and Grieser, 2007).

In this paper, we describe our study on the acoustic single bubbles in aqueous solutions of sodium dodecyl sulfate (SDS). SDS is one of the most common anionic surfactants, and its effects on the bubble growth rate and sonoluminescence have been studied in detail (Lee et al., 2005; Ashokkumar et al., 2007). Here we focus our attention to the stability conditions and the near-resonance dancing motion of bubbles, which are less studied but of practical importance to high-power and imaging ultrasound applications. We discuss our observations in relation to the previous studies on the SDS bubbles. As SDS is widely used in consumer chemicals for its bio-compatibility, our result may be useful in biomedical and clinical applications of acoustic bubbles in surfactant media.

MATERIALS AND METHODS

All chemicals used were of reagent grade. Solutions were prepared with highly deionized water from a reverse-osmosis and electro-deionization water purifier (Millipore Elix-3).

The experimental set-up (Figure 1) consisted of an electronic circuit and a 10-ml round-bottom flask, with a pair of torus-shaped ceramic piezoelectric transducers (Channel Industries C5800) as ultrasound actuators and a disc-shaped one (Channel Industries

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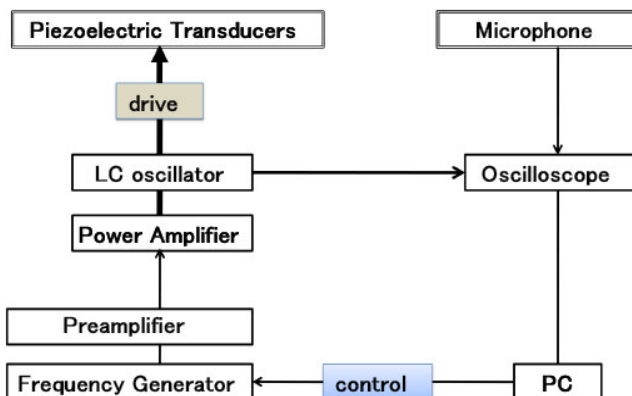
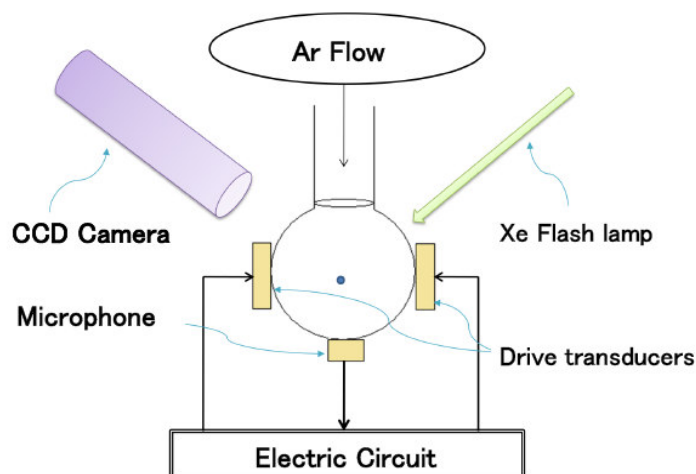


Figure 1. Experimental set-up: upper, the sample flask and ultrasonic transducers; lower, a schematic diagram of the drive electronics

C5400) as a microphone, both attached to the exterior glass surface of the flask (Putterman, 1995). The driving sinusoidal wave was generated in a home-built, direct-digital-synthesis frequency generator, amplified through a cascaded combination of two amplifiers and an LC oscillator, and then delivered to the actuators in phase. The driving frequency and voltage were controlled with a computer. The ultrasound was monitored with the microphone and an oscilloscope (Tektronix TDS2024).

Prior to each experiment, the flask was filled with an SDS solution, degassed, and purged with argon. A single bubble was either acoustically generated or induced by tapping with a capillary tube, and trapped approximately at the center of the flask by tuning the frequency and voltage to the resonant condition. The death of the bubble was clearly observed as a sudden disappearance of the bright spot in the illuminated volume. The lifetime of the bubble was evaluated by averaging the time from emergence/induction to disappearance over 10 samples for each condition. Images of the bubble were captured with a CCD camera (Philips PCVC840K; 15 frames/s) equipped with a zoom lens (Sigma AF28-80/3.5 - 5.6 Macro) by illuminating it with either a continuous Xe lamp (Hamamatsu L2274) or a Xe flashlamp (Hamamatsu L4643) via an optical fiber. In the latter case, the flashlamp was triggered with a

delay generator (SRS DG535) to deliver strobe pulses having a duration of 400 ns and synchronized to the CCD frame rate.

RESULTS AND DISCUSSION

The stability and lifetime of the acoustic bubbles show strong dependence on the ultrasound intensity and frequency, as well as the SDS concentration (Figure 2). It should be noted that important parameters such as the temperature and the dissolved gas concentration were not accurately controlled during the scanning of the driving amplitude or frequency, owing to the heat continuously evolved from ultrasound absorption; thus, the data from different scans often show discrepancy and should be taken as qualitative. At each SDS concentration, there is a well-defined maximum, that is, the optimal condition having the longest lifetime. At the constant frequency of $f = 74.5$ kHz, the bubble in pure

water lasts longest with the sonic amplitude $V = 550$ mV, whereas the condition shifts higher to $V = 700$ mV in the SDS solution of $c = 200$ μM .

Since the SDS concentrations were kept much lower than the critical micellar concentration ($\text{cmc} = 8$ mM), the SDS molecules in solution are expected to be in the forms of monomers and pre-micellar aggregates; they are thermodynamically unfavored and tend to populate at gas-water and glass-water interfaces. SDS-contaminated bubbles have a lower surface tension and a higher stability against dissolution, owing to protective organic shells. Accordingly, the observed extra stability of the SDS bubbles at a higher driving amplitude may not be surprising. On the other hand, effects of SDS on other macroscopic properties, such as the sound speed (Junquera, Peña and Aicart, 1994), shear viscosity (Kodama and Miura, 1972), and ultrasonic absorption (Yasunaga, Oguri and Miura, 1967), are all minor in dilute solutions below cmc . It is suggested, however, that SDS at low concentrations still affects surface viscoelastic properties, which may also contribute to the bubble growth acceleration, together with the effects on surface tension and interfacial mass transfer (Lee et al., 2005).

The acoustic field inside the round flask can be modeled by the forced radial pulsation of a liquid-filled, spherical enclosure wall (Blackstock, 2000). The lowest resonance frequency is given by $f_r = c_0 x'_{01} / 2\pi a$, where c_0 is the small-signal sound speed, $x'_{01} = 4.493$ is the first root (zero) of the derivative of the spherical Bessel function $J_0(x)$, and a is the flask radius. The associated standing pressure field has the antinode at the center of the flask ($r = 0$), and the node at the spherical surface of $r = \pi a / x'_{01} \sim 0.7 a$. The theoretical frequency, $f_r = 80.4$ kHz for water at 20°C , is close to but a little higher than our experimental resonance frequency range (74 - 75 kHz), possibly due to the deviation from spherical symmetry and the acoustic impedance mismatch of the glass-water interface.

This resonance frequency, in turn, determines the condition for a resonant bubble size (the equilibrium radius R_m) via the Minnaert theory of the pulsating spherical bubble: $f_r R_m = 3 \text{ m s}^{-1}$ in water (Leighton, 1997). Note that the theory is based on the balance between the internal gas pressure and the inertia of the moving liquid wall; the surface tension is assumed to be negligible. The resulting value in our case is $R_m = 40$ μm . The actual size of each bubble (R) is critical to its own fate: if $R < R_m$, the bubble travels to and stays at the antinode of the acoustic pressure field, driven by the acoustic radiation force (primary Bjerknes force); if $R > R_m$, it now travels to the node of the field and dissolves there; and if $R \sim R_m$, it tends to resonate violently, causing surface excitation, shape oscillation, dancing, or fragmentation.

Although the initial bubble size was not controlled in our experiments, those bubbles that were trapped at the flask center should have smaller radii than R_m . Such a bubble

is expected to grow during subsequent pulsations by a mechanism of the so-called rectified diffusion (RD), that is., active pumping of the dissolved gas into the interior of the bubble, provided that its size is above the RD threshold R_{RD} (Crum, 1980). It is often intuitively pictured that, after this growing, the bubble might quickly reach the Blake (or transient cavitation) threshold to undergo transient collapse, or reach R_m to undergo violent resonance and collapse. Detailed studies, however, have shown that it is by far more likely for the bubble to be trapped in a stable cyclic process of slight growth and shrinkage near the RD threshold, leading to the stable cavitation (Leighton, 1997).

Such stable bubbles typically survive over 10^5 cycles, and follow one of the two scenarios: if $R > R_{\text{RD}}$, it gradually grows via RD until R_m is reached, where violent oscillations and instabilities eventually result in the break up; otherwise, it stays at a relatively constant size and then dissolves away (Lee et al., 2005). In our experiments, we have observed both behaviors, the former being more frequent. Then, although the direct cause of the bubble death is unknown and probably statistical, we might expect the lifetime (t) to be inversely proportional to the RD growth rate (G): $t \sim (R_m - R)/G$. It turns out that this is not the case. The addition of SDS increases G (Lee et al., 2005), whereas t at its maximum is rather unchanged; the peak merely shifts to a higher driving amplitude (Figure 2). Accordingly, the bubble sustainability in the SDS solution seems to enjoy additional merits from interfacial stability and dynamic flexibility, in spite of the faster arrival to the resonance.

The distinction between 'stable' and 'transient' cavitation originates from numerical studies of the Rayleigh-Plesset equation. It becomes somewhat ambiguous in the case of repetitive sonoluminescence; the stable cavitation gains enough energy to attain temperatures similar to those in transient collapse, thus corresponding to the 'repetitive transient cavitation'. A suitable bubble size for sonoluminescence must be comparable with but a little smaller than R_m , so as to accept enough energy from the acoustic field, yet avoiding aspherical disturbance and energy loss by intensive resonance.

When the radius of the stable bubble approaches R_m , or the acoustic driving amplitude exceeds the stability limit, the bubble dynamics becomes complicated. Surface excitation and aspherical shape oscillation can be modeled by harmonic analysis if the deviation is small; otherwise, the bubble radius can lead to bifurcation and chaos. The position of the trapped bubble also starts fluctuating. Such positional 'dancing' motion is universally observed, but its experimental and theoretical understanding is far less, compared with other aspects of resonant bubble dynamics.

The CCD image of the bubble in our experiment reveals a significant dancing motion (Figure 3). The image (upper) under continuous illumination indicates the bubble size of 0.3 - 0.5 mm with 0.1 mm 'lumps'. This

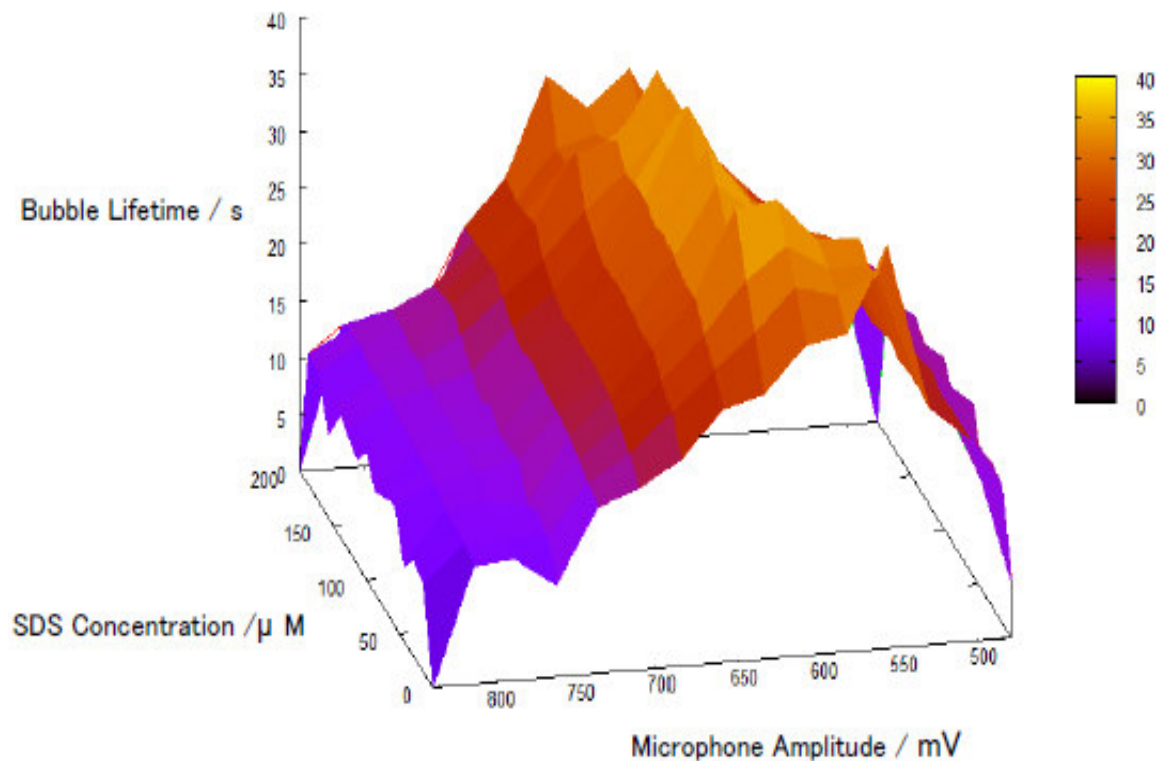


Figure 2. 3-Dimensional plots of the bubble stability. The average lifetime of single bubbles is shown as a function of the SDS concentration and the microphone amplitude (frequency = 74.5 kHz).

image is subject to the motional blur, as is clear from the 400 ns strobe image (lower) of the same bubble, where the bubble size is now reduced to 50 - 70 μm . The image, however, still shows blur, asphericity, and lumps of $\sim 10 \mu\text{m}$ (approaching the resolution limit). Whether it is still due to the motion, or to the intrinsic aspherical shape of the bubble, is unclear. In view of the resonance radius ($R_m = 40 \mu\text{m}$), our bubble is likely to be very close to the resonance, with intensive shape oscillations and deformation. It is possible that the radius may occasionally exceed R_m (which pops the bubble out of the antinode position), and then diminish below R_m (which drives it back into the antinode again).

The observed dancing motion is qualitatively similar in water and in the SDS solutions, although it is more pronounced in the latter. This is in contrast to the radial (expansion and contraction) dynamics of a single bubble under sonoluminescence conditions, where the effect of surfactants is insignificant (Ashokkumar et al., 2002). One possible explanation is the modification of acoustic streaming and microstreaming by SDS.

Both streaming and microstreaming are liquid flows under acoustic waves. The former results from absorption of acoustic momentum, whereas the latter is caused by local frictional forces, and confined to the acoustic microstreaming boundary layer of thickness $L_{ms} = (2\eta / \rho\omega)^{1/2}$, where η is the shear viscosity, ρ the density, and ω the circular frequency (Leighton, 1997). In our case, $L_{ms} =$

2 μm , thus microstreaming is below the CCD resolution limit and unlikely to be observed. On the other hand, streaming was observed when occasional dust particles approached the antinode. Effects of SDS on the streaming were, however, inconclusive.

The detailed trace of the bubble motion gives some insight into the dancing behavior (Figure 4). The bubble position (x, y) on each of the strobe CCD images has been determined by locating the maximum intensity over 5-pixel (up, down, left, right and middle) averages, and plotted three-dimensionally with the time course as the z-axis. The trace clearly shows a pendulum-like oscillatory motion, overshooting the antinode and coming back in the radial direction. As opposed to a pendulum, however, the inter-strobe average velocity of the bubble (arrow in the figure) tends to be lower in the vicinity of the antinode and higher in the periphery. The estimated velocity values are 0.07 - 1.2 mm/s, which represent lower bounds because of the two-dimensional projection to the CCD image plane. There also seems a periodic (modulated) variation in the amplitude envelope (that is, overshooting distance) and the direction of the pendulum motion. Such characteristics are intimately connected to the physical origin of the dancing motion, such as the Bjerknes forces and microstreaming, as well as to the possibility for chaos and sonoluminescence, so that a further study on the details will be intriguing.

In conclusion, we have found an enhanced driving



Figure 3. CCD Images of a bubble captured with the continuous Xe lamp (upper) or a single-shot of the Xe flashlamp (lower). The image width spans the actual scale of 3.27 mm.

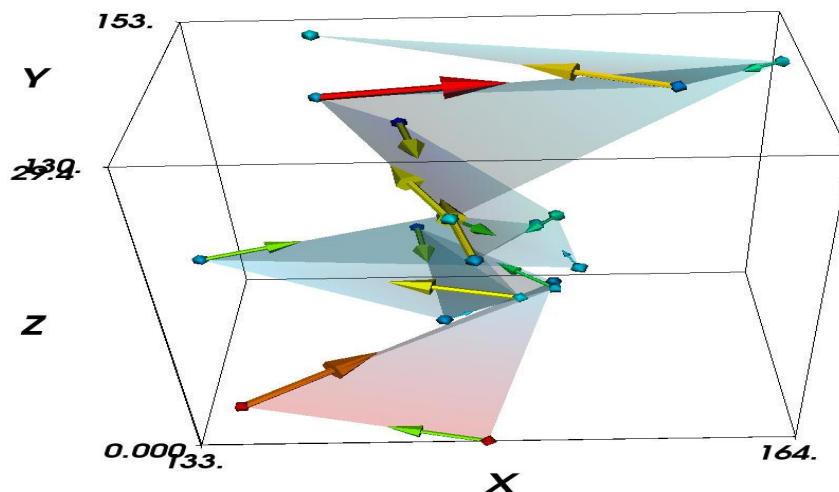


Figure 4. Dancing dynamics of the bubble. The bubble position on the CCD image (x , y) is plotted with the time course in the z direction (spanning 19 frames). The triangular strips depict each successive 3 positions for the aid to eyes; the arrows indicate the inter-frame average velocity of the translating bubble.

stability of acoustic single bubbles in dilute SDS solutions. We have also observed quasi-periodic traces of dancing motion of the bubble, which show an 'inverted pendulum' oscillation dynamics around the antinode.

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