Mechanisms for Stable Single Bubble Sonoluminescence

Michael P. Brenner,[1] Detlef Lohse,[2,3] David Oxtoby,[4] and Todd F. Dupont[3]

[1]*Department of Mathematics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*
[2]*Fachbereich Physik der Universität Marburg, Renthof 6, 35032 Marburg, Germany*
[3]*Department of Mathematics, The University of Chicago, Chicago, Illinois 60637*
[4]*Department of Chemistry and James Franck Institute, The University of Chicago, Chicago, Illinois 60637*

A gas bubble trapped in water by an oscillating acoustic field is expected to either shrink or grow on a diffusive time scale, depending on the forcing strength and the bubble size. At high ambient gas concentration this has long been observed. However, recent sonoluminescence experiments show that when the ambient gas concentration is low the bubble can be stable for days. This paper discusses mechanisms leading to stability.

Recent experiments on sonoluminescence (SL) [1–7] allow detailed studies of the dynamics of a bubble levitated in a periodic acoustic field. Besides the light emission itself, one of the greatest mysteries is how the bubble can exist in a stable state for many billions of cycles. Measurements of the time between successive light flashes show that the total mass of the bubble remains constant to high accuracy [1,2,7]. This result contradicts classical notions about the dynamics of periodically forced bubbles: An unforced bubble of ambient radius R_0 dissolves over a diffusive time scale, $\tau \sim \rho_0 R_0^2 / D(c_0 - c_\infty)$ [8], where ρ_0 is the ambient gas density in the bubble, D is the diffusion constant of the gas in the liquid, c_0 is the saturated concentration of the gas in the liquid, and c_∞ is the concentration of gas in the liquid far from the bubble. A strongly forced bubble grows by rectified diffusion, as first discovered by Blake [9,11]. This is because when the bubble radius is large, the gas pressure in the bubble is small, resulting in a strong mass flux into the bubble. Conversely, when the bubble radius is small there is a strong mass outflux. Since the diffusive time scale is much larger than the short time the bubble spends at small radii, gas cannot escape from the bubble during compression; the net effect is bubble growth. At a special value of the ambient radius R_0^* rectified diffusion and normal diffusion balance. However, the above arguments suggest that this equilibrium point is *unstable*; if the ambient radius is infinitesimally different from R_0^*, the bubble is pushed away from equilibrium. The classic papers on rectified diffusion (see, e.g., Eller and Crum [10–12]) verified the qualitative picture described above when $c_\infty/c_0 \approx 1$.

There are two controlled parameters in the SL experiments: the forcing pressure P_a and the gas concentration c_∞. The key to the discovery of *stable* single bubble SL (whose existence completely contradicts the above scenario) by Gaitan *et al.* [1] was that (i) $c_\infty/c_0 < 1$, and (ii) P_a must lie between a lower critical pressure and an upper critical pressure both of which depend on c_∞. Two different types of stable SL exist: In the first, the ambi-

ent radius remains constant for billions of cycles, as evidenced by the constant phase ϕ of the light emission relative to the oscillatory forcing. Barber and Putterman [2] showed that the "jiggle" in the phase differs by less than 50 psec from cycle to cycle. In the second type of stable SL, the bubble also persists for long periods although ϕ (and hence R_0) varies on a diffusive time scale [1,5]. The ambient radius grows until the bubble becomes parametrically unstable [5,13] and microbubbles pinch off. Experiments [5] show that this cycle can repeat indefinitely.

Transitions between the types of stable SL occur as a function of the gas concentration c_∞ as well as the forcing pressure. For pure argon bubbles when c_∞/c_0 is between approximately 0.06 and 0.25, ϕ oscillates on a diffusive time scale. At lower argon concentration $c_\infty/c_0 = 0.004$ the phase becomes perfectly stable [5].

Many of the dynamical phenomena exhibited by the SL experiments, including the existence of a stable bubble, might occur independently of the light emission. To study this question, we analyze the stability of a bubble with a simple model for the dynamics: although the model neglects many effects important for SL (including the light emission), it exhibits the qualitative features of both types of stable SL. When c_∞/c_0 is decreased at high enough forcing pressures, the classical unstable equilibrium point R_0^* undergoes a bifurcation and stabilizes [14]. In general, there can be *several* stable equilibria, although far from equilibrium small bubbles shrink and large bubbles grow. For higher P_a the window of stability closes, and the bubble can only survive through rectified diffusion followed by parametric instability. These results apply to any bubble oscillating at small enough c_∞/c_0, regardless of whether the forcing is strong enough to produce SL. Once the bubble enters the SL regime the simple theory might break down. For example, Löfstadt *et al.* suggest that nondiffusive effects [7] are necessary to account for stable air bubbles at strong forcing pressures. Our calculations of a dynamical model for specific nondiffusive effects show that they can indeed stabilize an unstable

equilibrium. The calculations suggest an experimental test to determine whether diffusive or nondiffusive effects dominate the SL experiments.

We first set up a formalism for studying the stability of the equilibrium point, following Fyrillas and Szeri [15] and Löfstedt et al. [7]. Let $c(r, t)$ denote the concentration of gas dissolved in the liquid a distance r from the center of the bubble. For $r > R(t)$, where $R(t)$ is the radius of the bubble, c satisfies a convection diffusion equation

$$\partial_t c + \frac{R^2 \dot{R}}{r^2} \partial_r c = D \nabla^2 c . \tag{1}$$

The boundary conditions are given by Henry's law $c(R, t) = c_0 P(R, t)/P_0$ and by $c(\infty, t) = c_\infty$. The concentration gradient at the boundary gives the mass loss/gain of the bubble $\dot{M} = 4\pi R^2 D \partial_r c|_{R(t)}$.

These equations determine the growth of the bubble as a function of time. There are two crucial observations: First, Eller noted that changing coordinates to $h = (r^3 - R^3)/3$ and $\tau = \int^t R^4 \, dt$ transfers Eq. (1) to the simpler form

$$\partial_\tau c = D \partial_h \left[\left(1 + \frac{3h}{R^3} \right)^{4/3} \partial_h c \right] = 0 . \tag{2}$$

For the following it is convenient to define the τ average of a function $f(t)$ by $\langle f(t) \rangle_\tau = \int f(t) R(t)^4 \, dt / \int R(t)^4 \, dt$.

The second observation [7,15] is that the bubble radius changes over a much faster time scale than the ambient radius. Averaging Eq. (2) over the fast time scale gives the dynamics of the ambient radius,

$$\rho_0 R_0^2 \frac{dR_0}{d\tau} = D \frac{c_\infty - \langle c(R(t), R_0(t)) \rangle_\tau}{\int_0^\infty dh / \langle 1 + 3h/R^3 \rangle_\tau} . \tag{3}$$

Equilibrium points satisfy

$$\frac{\langle p \rangle_\tau}{P_0} = \frac{c_\infty}{c_0} . \tag{4}$$

The equilibrium is stable if the quantity $\beta = d\langle p \rangle_\tau / dR_0$ is positive.

Now we proceed to analyze this model. We calculate numerically $\langle p \rangle_\tau$ as a function of R_0, for different driving pressures, by τ averaging solutions $R(t)$ of the Rayleigh-Plesset (RP) equation. The RP equation [6,16,17] governs the dynamics of an acoustically forced bubble, and is given by

$$R\ddot{R} + \frac{3}{2} \dot{R}^2 = \frac{1}{\rho_w} [p(R, t) - P(t) - P_0] + \frac{R}{\rho_w c_w}$$
$$\times \frac{d}{dt} [p(R, t) - P(t)] - 4\nu \frac{\dot{R}}{R} - \frac{2\sigma}{\rho_w R} . \tag{5}$$

We use parameters corresponding to [3,6,18] an air bubble in water: the surface tension of the air-water interface is $\sigma = 0.073$ kg/s^2, while water has viscosity $\nu = 10^{-6}$ m^2/s, density $\rho_w = 1000$ kg/m^3, and speed of sound $c_w = 1481$ m/s. The acoustic field is driven via $P(t) = P_a \cos(\omega t)$ with $\omega/2\pi = 26.4$ kHz and external

pressure $P_0 = 1$ atm. The pressure inside the bubble varies adiabatically like $p(R) \sim (R^3 - a^3)^{-1.4}$. Here $a = R_0/8.73$ is the hard core van der Waals radius.

Figure 1 shows $\langle p \rangle_\tau / P_0$ for several values of P_a. For small P_a, $\langle p \rangle_\tau$ monotonically decays with R_0, signaling a diffusively unstable equilibrium. For example, when $c_\infty/c_0 = 1$ with a forcing amplitude of $P_a = 0.8$ the unstable equilibrium occurs at $R_0 \approx 5 \mu$m. Note that at large R_0 the bubble becomes unstable to shape oscillations [13].

At large P_a, however, $\langle p \rangle_\tau$ develops oscillations as a function of R_0, so for a range of c_∞/c_0 there are several stable equilibrium points. As an example, see the inset of Fig. 1: when $c_\infty/c_0 = 10^{-2}$ and $P_a = 1.25$ atm, there are stable equilibria (denoted by small dots in the figure) at $R_0 = 6.5, 6.8, 7.1, 7.5, 8.0$, and 8.5μm. To further verify the existence of multiple stable equilibria, we have solved the full equations (1) and (5) numerically with a standard finite difference scheme. Indeed, the ambient radius saturates at different values, depending on the initial bubble size. These equilibria are approached both from above and from below. Details will be published elsewhere [19].

We now outline the predictions of these calculations for experiments. A common protocol in the SL experiments [1,3] is to slowly increase the driving pressure P_a. The initial ambient radius depends on the preparation of the bubble. At low pressures the bubble shrinks, since $c_\infty < \langle c \rangle_\tau$. As the forcing pressure is increased, there is a critical pressure where equilibrium points (both stable and unstable) appear in the parametrically stable region [20]: calculations for $c_\infty/c_0 = 0.25$ indicate this occurs near 1 atm. Above this forcing pressure, the bubble can follow
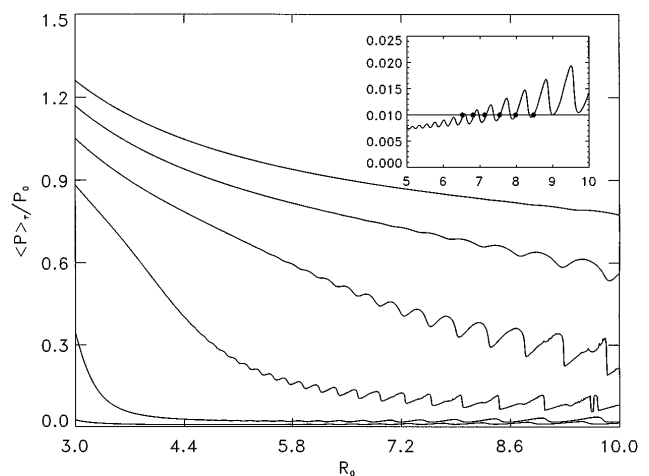

FIG. 1. $\langle p \rangle_\tau / P_0$ as a function of R_0 (in μm) for $P_a = 0.8$, 0.9, 1, 1.1, 1.2, and 1.25 atm, top to bottom. Equilibrium corresponds to $\langle p \rangle_\tau / P_0 = c_\infty/c_0$. The equilibrium is diffusively stable if the slope $\beta = d\langle p \rangle_\tau / dR_0$ is positive. Inset: An enlargement of $P_a = 1.25$ atm. The straight line corresponds to $c_\infty/c_0 = 10^{-2}$. The intersection of the straight line with the curve corresponds to equilibrium points. When $\beta > 0$ (the solid dots in the figure) the equilibrium is stable.

three different scenarios, depending on its ambient radius R_0: (a) If R_0 is smaller than all equilibrium radii, the bubble shrinks. (b) If R_0 is near a stable equilibrium, the bubble is attracted to it and thus maintains a constant ambient radius thereafter. (c) If R_0 is larger than all the equilibrium radii, the bubble grows by rectified diffusion. The particular scenario that occurs depends on the initial R_0 and so could vary from experiment to experiment. Also note that the system is hysteretic: the sequence of states occurring when the forcing pressure increases will not, in general, be repeated when the forcing pressure decreases. In the experiments of Barber et al. [3], the bubble initially follows (c), growing by rectified diffusion. When the ambient radius becomes large the bubble is parametrically unstable; the bubble can decrease its radius by pinching off a microbubble. As the forcing pressure is further increased, the bubble continues diffusive growth followed by microbubble pinchoff; the ambient radius is thus controlled by the parametric instability line [13]. Eventually, for even larger P_a, microbubble pinchoff is severe enough to place the ambient radius near a stable equilibrium, after which the bubble follows scenario (b). Note that this transition from a "jiggling" bubble to a stable bubble should always be accompanied by a discontinuous jump in the ambient radius. Such jumps are observed by Barber et al. [3] for air bubbles near the onset of SL. For $c_\infty/c_0 = 0.25$ the stable state persists until $P_a \approx 1.15$; above $P_a = 1.15$ within the simple model the equilibrium point destabilizes. The bubble must return to diffusive growth followed by microbubble pinching to survive. At even larger P_a all parametrically stable bubbles shrink, so the continued existence of a stable bubble is impossible. The sequence of events predicted by the simplified model is qualitatively similar to those in the SL experiments.

The stable equilibrium points are ultimately due to oscillations in $\langle p \rangle_\tau$ as a function of R_0, which arise from resonances in the Rayleigh-Plesset equation. Oscillations even occur in the maximum radius as a function of R_0, so that in some situations adding more gas to a bubble decreases its maximum size. A comparison with the Mathieu equation is instructive: If the eigenfrequency (in the RP equation this depends on R_0) is an integer or half integer fraction of the forcing frequency, the amplitude of the oscillations is anomalously large. A detailed study is in progress [19].

Quantitative agreement between the simple model and the SL experiments requires accounting for several neglected effects. These include realistic heat transfer [6,21,22] and equations of state for the gas [23], as well as spatial variations of the pressure within the bubble [23,24]. To illustrate the dependence of solutions of the RP equation on material parameters, consider a 3.3 μm bubble at forcing pressure $P_a = 1.3$ atm. Upon changing $\gamma = 1$ (isothermal) to $\gamma = 1.4$ (adiabatic) the maximum radius R_{max} changes by 20%; setting the surface tension to zero changes R_{max} by 50%; changing the fluid viscosity

from water to 0.07 cm^2/sec changes R_{max} by 30%. Increasing the forcing pressure by 0.05 atm (the error in experiments [3]) increases R_{max} by 20%. The uncertainty in the effective values of all the aforementioned quantities during SL translates into uncertainties in predicted positions for the stable equilibria. However, our qualitative conclusions are robust, occurring throughout the relevant range of parameter space.

Multiple stable equilibria exist for a range of forcing pressures for any $c_\infty/c_0 \ll 1$; although these states generally occur at high forcing, the bubble oscillations need not be strong enough to produce light [25]. What parameter regime corresponds to SL in the simplified model? No established criterion exists, though experiments [3,26] suggest that the collapse ratio R_{max}/R_0 is the relevant parameter. For $c_\infty/c_0 = 0.2$, the stable equilibria have collapse ratios $R_{max}/R_0 \approx 3$; for $c_\infty/c_0 = 0.004$, $R_{max}/R_0 \approx 7$. The former c_∞/c_0 corresponds to the strongest SL for air bubbles; the latter corresponds to the new phase (i.e., the low c_∞ phase) of SL recently discovered [5] for argon bubbles. The largest discrepancy between this simple model and SL experiments is its inability to explain why the strongest light emission occurs at a much larger c_∞/c_0 in air bubbles than argon bubbles.

This discrepancy is the basic reason for Löfstadt et al.'s speculation that "nondiffusive" effects may be necessary to explain stable SL in air. We have verified through dynamical calculations that a simple nondiffusive effect can stabilize an otherwise unstable equilibrium point [27]: the increase of the mass diffusion constant [D in Eq. (3)] with temperature and pressure. When the bubble pressure and temperature is high, the diffusion constant near the bubble wall is larger than the diffusion constant in the bulk liquid, leading to additional mass outflux from the bubble.

This increase in the interfacial diffusion constant can be studied with a simple model: Whenever the pressure inside the bubble exceeds a critical pressure p_{thres} we discontinuously increase the diffusion constant near the bubble wall by a factor f_{thres}. The diffusion constant in the bulk liquid remains constant, since high pressures and temperatures are localized near the bubble wall. Numerical simulations of the full equations [19] with and without this effect demonstrate that the unstable equilibrium point can be stabilized for a wide range of p_{thres} and f_{thres}. The position of the equilibrium point is shifted to larger radii, as predicted by Löfstadt et al. [7].

A central question for both theory and experiment is determining the parameter ranges where diffusive effects alone produce a stable bubble. The present results suggest that the qualitative features of the bubble dynamics in SL experiments also arise within classical theories of bubble dynamics. Answering definitively whether novel effects are needed to explain stable SL is complicated by modeling uncertainties. The present calculations provide qualitative criteria to assist in answering this question: although both diffusive and nondiffusive effects can lead to

stable equilibria, only the diffusive effects lead to discrete equilibria. In order to determine which effect dominates the SL experiments, we suggest that experiments search for the discretization of the ambient radius in both light emitting and nonlight emitting bubbles.

Note added.—During the review process, we became aware of a conference proceeding [28] by Crum and Cordry who reached similar conclusions about the existence and possible importance of multiple stable equilibrium points. These authors also presented preliminary experimental evidence for the discrete equilibria.

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