The Role of Surface Tension in Stable Single-Bubble Sonoluminescence

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(Received 26 June 1996; revised manuscript received 12 November 1996)

A theory for stable bubble oscillations in high pressure sound fields is presented. It is based on the strong influence of the surface tension on the dynamics of small bubbles and takes into account rectified diffusion and the resonancelike response of small bubbles to very strong acoustic pressure amplitudes. This theory provides an explanation for the existence of small, stably oscillating bubbles that have been observed in experiments on sonoluminescence. [S0031-9007(96)02169-2]

PACS numbers: 43.25.+y, 43.35.+d, 47.55.Bx, 78.60.Mq

Bubbles in a liquid that are subject to an external sound field not only oscillate strongly nonlinearly but may also emit light. This phenomenon is called *sonoluminescence* (SL) and was discovered by Marinesco and Trillat [1] in 1933. Since then it has been investigated experimentally as well as theoretically by many authors [2–12]. The interest in SL was restimulated by the elaborate experiments of Gaitan *et al.* [6], who investigated SL of a single bubble in water trapped by a strong periodical acoustic field. This phenomenon is called *single-bubble sonoluminescence* (SBSL) and was investigated in a number of papers [7–11].

One of the most striking results of SBSL experiments was the observation that bubbles can oscillate permanently for several days without dissolution and without changing their size. A detailed investigation of the underlying physical mechanism shows that there are many effects and phenomena that have to be taken into account. This list includes rectified diffusion, surface tension, dissolved gas, thermoconductivity, acoustic radiation, viscosity, microstreaming around a bubble (maybe generated by surface waves on the bubble [13]), the nonlinear character of bubble oscillations, and the fragmentation and coalescence of bubbles [14–17]. The analysis of experimental and theoretical results shows that the main effects that are important for the generation of stable cavitation bubbles are rectified diffusion, surface tension, and shape oscillations. The mechanisms of how gas diffusion may lead to stability of the bubble size were discussed in [18] in order to analyze the possibility of multiple stable equilibrium radii of the bubble. The stability of shape oscillations was investigated in [19,20] where it was shown that large bubbles have a tendency to disintegrate due to unstable surface oscillations. In this Letter we focus on the influence of rectified diffusion and surface tension on the stability of small bubbles in a sound field.

Without external sound field, bubbles of any size are unstable because the pressure inside the bubble is larger than in the liquid, and therefore the bubble will dissolve slowly due to a continuous mass flux from the interior of the bubble into the liquid. In the presence of a periodic acoustic field the bubble starts to oscillate. During the expansion period gas diffuses from the liquid into the bubble, and during the contraction cycle the diffusion process takes place in the opposite direction. There is a net flow of gas into the bubble because the area of the bubble wall is greater during the expansion period and therefore more gas will enter during the expansion than will leave during the contraction cycle. This phenomenon is called *rectified diffusion* and leads to a growth of the bubble [21,22]. For small amplitudes it was shown [23,24] that the growth rate depends on the sound field amplitude P_a, the resonance radius R_r, and the equilibrium radius R_0 of the bubble. *This growth rate is closely correlated with the response curve that describes the dependence of the maximum bubble radius on the equilibrium radius.* The theory for weakly nonlinear oscillations gives a good description of the growth and dissolution processes of sufficiently large bubbles and small pressure amplitudes. However, it provides no answer to the questions pertaining to (stable) sonoluminescence: *Why are small gas bubbles in a liquid stable in the presence of a strong sound field?*

For single-bubble oscillations under medium and large pressure amplitudes a complicated scenario of bifurcations and coexisting (chaotic) attractors exists [25,26]. Our numerical simulations show, however, that for very small bubbles in very strong sound fields the dynamics becomes more regular and a new type of strong resonance with a thresholdlike increase in oscillation amplitude occurs [12,27]. The physical reason for this phenomenon is the fact that for very small bubbles the surface tension pressure $P_\sigma = 2\sigma/R_0$ is very high and the bubbles behave like flexible solid particles even for large driving pressures. One cycle of a typical bubble oscillation for this case is shown in Fig. 1(b) for the normalized bubble radius $R(t)/R_0$. Figure 1(a) shows the driving pressure of the external sound field $p_a(t) = -P_a \sin(\omega t)$. When

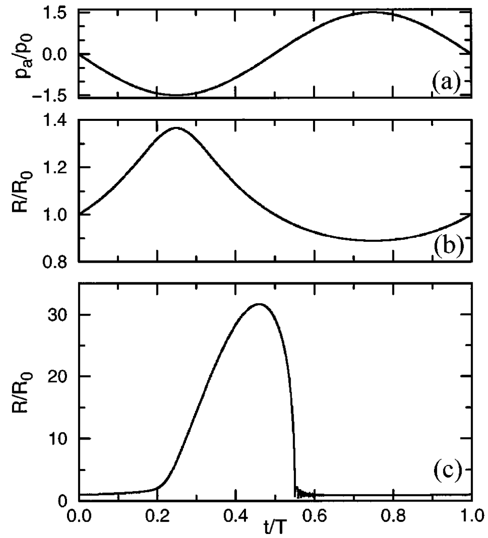

FIG. 1. The influence of surface tension on bubble oscillations. Plotted is one period of oscillation (frequency $\nu = \omega/2\pi = 20$ kHz) of the normalized sound field pressure $p_a(t)/p_0$ (a) and of the normalized bubble radius $R(t)/R_0$ vs time t (b), (c) for a pressure amplitude of $P_a = 1.5$ bar. (b) A bubble with equilibrium radius $R_0 = 1$ μm oscillates with small amplitude due to the relatively high surface tension pressure $P_\sigma = 2\sigma/R_0$. (c) For $R_0 = 1.5$ μm strongly nonlinear oscillations with high amplitude occur.

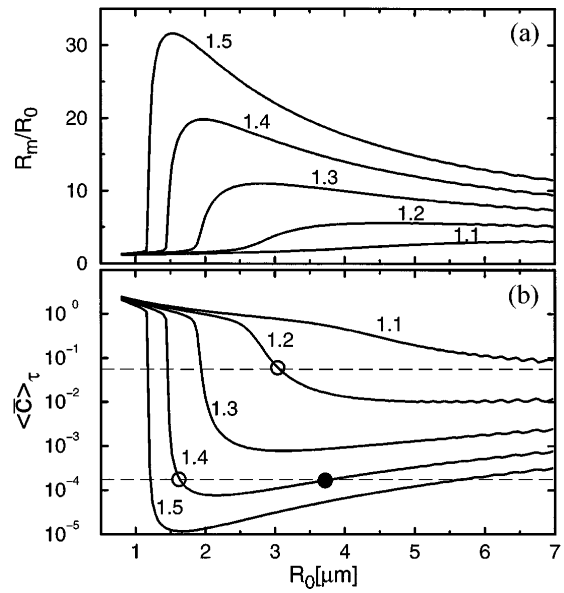

FIG. 2. (a) Response curves showing the normalized maximum bubble radius R_m/R_0 vs the equilibrium radius R_0 for different pressure amplitudes $P_a = 1.1-1.5$ bar. (b) Illustration of the cavitation threshold showing the nonlinearly averaged concentration near the bubble wall $\langle \bar{c} \rangle_\tau$ vs the equilibrium radius R_0 for different pressure amplitudes $P_a = 1.1-1.5$ bar. The horizontal dashed lines denote different levels of gas concentrations $\bar{c}_\infty$ in the liquid. The open circles denote the cavitation threshold value R_{th} of the radius and the filled circle the stable bubble radius R_s.

we increase the size of the bubble R_0 it starts to oscillate differently. During the expansion period the influence of the surface tension decreases rapidly, and therefore the amplitude of the expansion grows enormously leading to a strong collapse. The transition point may be called *nonstatic Blake threshold* [3,28]. The kind of oscillation present beyond this threshold is shown in Fig. 1(c). The results given in Fig. 1 and in all following figures have been computed using the Keller-Miksis model [26,29]:

$$\left(1 - \frac{\dot{R}}{C_l}\right) R\ddot{R} + \frac{3}{2}\dot{R}^2\left(1 - \frac{\dot{R}}{3C_l}\right) = \left(1 + \frac{\dot{R}}{C_l}\right)\frac{P}{\rho} + \frac{R}{\rho C_l}\frac{dP}{dt},$$

with

$$P = \left(p_0 + \frac{2\sigma}{R_0}\right)\left(\frac{R_0}{R}\right)^{3\kappa} - p_0 - \frac{2\sigma}{R} - \frac{4\mu}{R}\dot{R} - p_a(t),$$

for air bubbles in water at 20° C with $\kappa = 1.4$, $\sigma = 0.0725$ N/m, $p_0 = 1$ bar, $C_l = 1500$ m/s, and a driving frequency of $\omega = 2\pi \cdot 20$ kHz. Qualitatively the same results have been obtained for the Gilmore model [30].

When the equilibrium radius of the bubble is increased further the influence of the surface tension pressure P_σ becomes smaller and a nonmonotonous resonance curve for the normalized radius R_m/R_0 occurs. These response curves are shown in Fig. 2(a) for different values of the pressure P_a. One can see that the nonmonotonous behavior starts for $P_a > 1.2$ bar.

Now we investigate the rectified diffusion in the strong resonance region for small bubbles. The theoretical formulation for mass transport across the dynamic interface associated with a spherical bubble undergoing volume oscillations was derived in Ref. [31]. The equations governing the convection and diffusion of dissolved gas in the liquid outside a spherical bubble can been written in the following form:

$$\frac{\partial c}{\partial t} + \frac{R^2(t)\dot{R}(t)}{r^2}\frac{\partial c}{\partial r} = \frac{D}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial c}{\partial r}\right), \tag{1}$$

$$c|_{r=R(t)} = H\left(p_0 + \frac{2\sigma}{R_0}\right)\left(\frac{R(t)}{R_0}\right)^{-3\kappa}, \qquad c|_{r=\infty} = c_\infty, \tag{2}$$

$$\frac{dm}{dt} = 4\pi R^2(t) D \left.\frac{\partial c}{\partial r}\right|_{r=R(t)}. \tag{3}$$

Here Eq. (1) describes the convective diffusion, where $R(t)$ is the bubble radius governed by some dynamical equation for bubble oscillations, c is the mass concentration of gas dissolved in the liquid, and D is the diffusivity of the gas in the liquid. Equation (2) gives the boundary conditions at the bubble surface and far from it provided by Henry's law, which relates the concentration of gas in a liquid to the partial pressure of the gas above the liquid. The symbol H denotes Henry's constant and c_∞ is the initial uniform gas concentration in the fluid where the bubble is assumed to be created. Equation (3) describes the rate of the gas transport across the bubble interface.

This mass transport problem was solved approximately for large Peclet numbers [31] ($\mathrm{Pe} = R_0^2\omega/D \gg 1$). In this case Eqs. (1)–(3) can be simplified by a transformation of the problem into normalized Lagrangian coordinates $\eta = [r^3 - R^3(t)]/3R_0^3$ to avoid difficulties because of the moving boundary conditions. Another analytical difficulty—oscillatory behavior of the concentration close to the bubble surface and slow diffusion behavior farther away from the bubble—was solved by splitting the problem into two parts. Finally, it was shown that the time averaged rate of mass transport in the case of any nonlinear periodic bubble oscillation may be approximated as follows:

$$\frac{d\bar{m}}{d\tau} = \frac{\bar{c}_\infty - \langle \bar{c} \rangle_\tau}{T_{rd}}, \quad T_{rd} = \int_0^\infty \frac{d\eta}{\langle [3\eta + \bar{R}^3(t)]^{4/3} \rangle}, \tag{4}$$

$$\bar{c} = \frac{c(R(t),t)}{T_{rd}}, \quad \bar{c}_\infty = \frac{c_\infty}{c_0}, \quad \bar{m} = \frac{m}{m_0}, \quad \bar{R} = \frac{R}{R_0}.$$

Here c_0 is the saturation concentration in the liquid separated from gas at pressure p_0 by a plane boundary and m_0 is the mass of the gas soluted in the liquid displaced by the undisturbed bubble. The variable $\tau = tD/R_0^2$ is the slow diffusion time scale, and T_{rd} is the dimensionless characteristic time of rectified diffusion mass growth rate of the bubble.

In this approach two different averaging procedures are used. First, ordinary averaging over the period T of the acoustic field

$$\langle f(t) \rangle = \frac{1}{T} \int_0^T f(t)\, dt\,;$$

and second, a "nonlinear averaging" procedure in a specific nonlinear time scale

$$\langle f(t) \rangle_\tau = \frac{1}{\int_0^T R^4(t)\, dt} \int_0^T R^4(t) f(t)\, dt\,,$$

where the radius evolution $R(t)$ is used for computing the gas concentration near the bubble wall $\langle \bar{c} \rangle_\tau$. This approach was also used by Brenner *et al.* [18] to investigate the multiple stable equilibrium radii of the bubble for medium pressure amplitudes. Here we use it to consider the stability problem in the case of very small bubbles in a strong acoustic field.

Figure 2(b) shows the averaged gas concentration $\langle \bar{c} \rangle_\tau$ near the bubble wall vs the equilibrium radius R_0 of the bubble for different amplitudes P_a of the acoustic field. For small and medium values of P_a the concentration decreases monotonically as a function of the equilibrium radius. For sufficiently large amplitudes P_a, however, the corresponding concentration curves possess a global minimum for small bubble radii. This nonmonotonous dependence of $\langle \bar{c} \rangle_\tau$ on R_0 is a result of the strong resonance shown in Fig. 2(a).

Since the characteristic time T_{rd} in Eq. (4) is always positive the evolution of the mean bubble mass depends only on the difference between the concentration of gas in the liquid $\bar{c}_\infty$ and the nonlinearly averaged concentration near the bubble wall $\langle \bar{c} \rangle_\tau$. As can be seen in Fig. 2(b) there are two possible scenarios. If $\bar{c}_\infty$ is large [upper dashed line in Fig. 2(b)] a single equilibrium point $\langle \bar{c} \rangle_\tau = \bar{c}_\infty$ exists that is unstable. This case is denoted in Fig. 2(b) by the open circle at the point of intersection of the upper dashed line with the concentration curve for $P_a = 1.2$ bar. The unstable equilibrium provides a threshold value R_{th} for the bubble radius. Bubbles with radius $R_0 < R_{\mathrm{th}}$ dissolve due to diffusion flux from the bubble into the liquid. On the other hand, bubbles with $R_0 > R_{\mathrm{th}}$ grow permanently due to rectified diffusion until they become very large (and may disintegrate).

If $\bar{c}_\infty$ is small [lower dashed line in Fig. 2(b)] two equilibrium points $\langle \bar{c} \rangle_\tau = \bar{c}_\infty$ exist. The left fixed point denoted by the open circle in Fig. 2(b) is unstable and closely related to the previous case. It defines the cavitation threshold radius R_{th}. The equilibrium point plotted as a filled circle at the right hand side in Fig. 2(b) is stable and provides a stable radius R_s for single bubbles oscillating in the acoustic field. Bubbles with radius $R_{\mathrm{th}} < R < R_s$ grow until they reach the stable radius R_s. If the bubble radius is larger than R_s the bubble shrinks until $R_0 = R_s$. A necessary condition for the existence of R_s is a nonmonotonous dependence of the nonlinearly averaged concentration $\langle \bar{c} \rangle_\tau$ on the equilibrium radius R_0 for small bubbles. Only in this case a range of $\bar{c}_\infty$ values exists such that stable bubble oscillations are possible. For given small values of the concentration of gas in the liquid $\bar{c}_\infty$ there exists a lower threshold value P_a^c for the pressure amplitudes P_a that leads to stable bubbles as can be seen in Fig. 3 showing the dependence of the threshold value R_{th} (dashed curve) and the stable bubble radius R_s (solid curve) on the pressure amplitude P_a. For small values of P_a bubbles of any size will dissolve. At some critical value P_a^c the stable bubble radius R_s occurs due to a saddle-node bifurcation. When the pressure P_a is increased further the value of R_s increases, the bubble becomes very large and will eventually be destroyed due to dynamical instabilities (e.g., surface oscillations). Such a finite pressure range for stable bubble oscillations has also been observed experimentally [6].

In this Letter a theory for stable bubble oscillations in high pressure sound fields has been presented that is based on the strong influence of the surface tension on the dynamics of small bubbles. The approach presented takes into account the interaction of two effects: rectified diffusion and the resonancelike response of small bubbles on

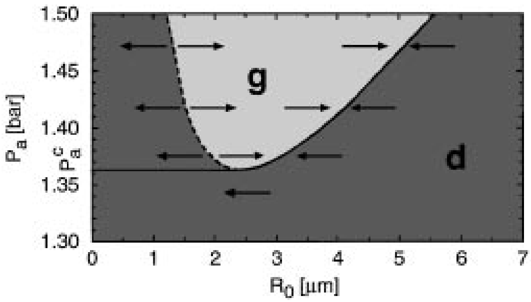

FIG. 3. Cavitation threshold radius R_{th} (dashed curve) and stable bubble radius R_s (solid curve) in dependence on the pressure amplitude P_a for fixed small value of the gas concentration $\bar{c}_\infty$ corresponding to the lower dashed line in Fig. 2(b). The islands of growth (light shading) in the sea of dissolution (dark shading) is a necessary prerequisite for stable bubble oscillations and thus SBSL to occur.

very strong acoustic pressure amplitudes due to the surface tension pressure. The results provide an explanation for the existence of small, stably oscillating bubbles that have been observed in experiments on sonoluminescence.

During the reviewing process a paper appeared where the occurrence of sonoluminescence in the parameter space (R_0, P_a) has been determined experimentally for just the case studied here, the small bubble, large acoustic pressure amplitude limit [32]. There one main finding is that sonoluminescence experimentally occurs along a *line* in the (R_0, P_a) parameter space, just as predicted by our Fig. 3 (solid line). We predict and with the measurement technique of Holt and Gaitan [32] it can be checked that this line starts at a bifurcation value where the stable and unstable branch of bubble oscillations equilibrium meet opening up an island of growth in the sea of dissolution. Both theory and experiment fall short in explaining the low gas content in the bubble. This is not at all astonishing as a wealth of other phenomena are not considered, for instance pertaining to the chemistry inside the bubble.

This work was supported by the *Projektträgerschaft Internationales Büro* (Contract No. X222.31), the Ministry for Science, Higher Education and Technical Policy of the Russian Federation and the State Committee of the Russian Federation for Higher Education.

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[1] N. Marinesco and J. J. Trillat, Proc. R. Acad. Sci. **196**, 858 (1933).
[2] H. Kuttruff, Acustica **12**, 230 (1962).
[3] A. J. Walton and G. T. Reynolds, Adv. Phys. **33**, 595 (1984).
[4] K. S. Suslick, Science **247**, 1439 (1990).
[5] B. P. Barber and S. J. Putterman, Nature (London) **352**, 318 (1991).
[6] D. F. Gaitan, L. A. Crum, C. C. Church, and R. A. Roy, J. Acoust. Soc. Am. **91**, 3166 (1992).
[7] R. Löfstedt, B. P. Barber, and S. Putterman, Phys. Fluids A **5**, 2911 (1993).
[8] R. G. Holt, D. F. Gaitan, A. A. Atchley, and J. Holzfuss, Phys. Rev. Lett. **72**, 1376 (1994).
[9] L. A. Crum, Phys. Today **47**, 22 (1994); L. A. Crum and S. Cordry, in *Bubble Dynamics and Interface Phenomena*, edited by J. R. Blake *et al.* (Kluwer, Dordrecht, 1994).
[10] B. P. Barber, K. Weninger, R. Löfstedt, and S. Putterman, Phys. Rev. Lett. **74**, 5276 (1995).
[11] R. Löfstedt, K. Weninger, S. Putterman, and B. P. Barber, Phys. Rev. E **51**, 4400 (1995).
[12] C. D. Ohl, H.-C. Nägerl, R. Blatt, and W. Lauterborn, *DAGA—Fortschritte der Akustik*, edited by T. Portele and W. Hess (DEAG e.V., Oldenburg, 1996), pp. 418–419.
[13] C. C. Church, J. Acoust. Soc. Am. **83**, 2210 (1988).
[14] *Cavitation and Inhomogeneities in Underwater Acoustics*, edited by W. Lauterborn (Springer, Berlin, 1980).
[15] F. R. Young, *Cavitation* (McGraw-Hill Book Company, London, 1989).
[16] T. G. Leighton, *The Acoustic Bubble* (Academic Press, London, 1994).
[17] R. I. Nigmatulin, *Dynamics of Multiphase Media* (Hemisphere Publishing Corporation, New York, 1991), Vols. 1,2.
[18] M. P. Brenner, D. Lohse, D. Oxtoby, and T. F. Dupont, Phys. Rev. Lett. **76**, 1158 (1996).
[19] H. W. Strube, Acustica **25**, 289 (1971).
[20] M. P. Brenner, D. Lohse, and T. F. Dupont, Phys. Rev. Lett. **75**, 954 (1995).
[21] D.-Y. Hsieh and M. S. Plesset, J. Acoust. Soc. Am. **33**, 206 (1961).
[22] A. Eller and H. G. Flynn, J. Acoust. Soc. Am. **37**, 493 (1963).
[23] L. A. Crum and G. M. Hansen, J. Acoust. Soc. Am. **72**, 1586 (1982).
[24] N. A. Gumerov, Prikl. Mat. Mekh. **55**, 2256 (1991).
[25] W. Lauterborn, J. Acoust. Soc. Am. **59**, 283 (1976).
[26] U. Parlitz, V. Englisch, C. Scheffczyk, and W. Lauterborn, J. Acoust. Soc. Am. **88**, 1061 (1990).
[27] W. Lauterborn, Acustica **20**, 105 (1968).
[28] F. G. Blake, Technical Memo 12, Acoustics Research Laboratory, Harvard University, 1949.
[29] J. B. Keller and M. Miksis, J. Acoust. Soc. Am. **68**, 628 (1980).
[30] F. R. Gilmore, Hydrodynamics Laboratory, California Institute of Technology, Pasadena, California, Report No. 26-4, 1952.
[31] M. M. Fyrillas and A. J. Szeri, J. Fluid Mech. **277**, 381 (1994).
[32] R. G. Holt and D. F. Gaitan, Phys. Rev. Lett. **77**, 3791 (1996).