

Boosting Sonoluminescence

Joachim Holzfuss¹, Matthias Rüggeberg¹, Robert Mettin^{1,2}

¹*Institut für Angewandte Physik, TU Darmstadt, Schlossgartenstr. 7, 64289 Darmstadt, Germany*

²*Drittes Physikalisches Institut, Bürgerstr. 42-44, 37073 Göttingen, Germany*

(Accepted for publication in Phys. Rev. Lett.)

(Copyright 1998 by The American Physical Society)

Single bubble sonoluminescence has been experimentally produced through a novel approach of optimized sound excitation. A driving consisting of a first and second harmonic with selected amplitudes and relative phase results in an increase of light emission compared to sinusoidal driving. We achieved a raise of the maximum photo current of up to 300% with the two-mode sound signal. Numerical simulations of multimode excitation of a single bubble are compared to this result.

PACS numbers: 78.60.Mq, 42.65.Re, 43.25.+y

By focusing ultrasonic waves of high intensity into a liquid, thousands of tiny bubbles appear. This process of breakup of the liquid is called acoustic cavitation. The bubbles begin to form a fractal structure that is dynamically changing in time. They also emit a loud chaotic sound because of their forced nonlinear oscillations in the sound field [1]. The large mechanical forces on objects brought into contact with the bubbles enable the usage of cavitation in cleaning, particle destruction and chemistry. Marinesco and Trillat [2] found that a photo plate in water could be fogged by ultrasound. This multi-bubble sonoluminescence (MBSL) has been analyzed by many researchers, and a great amount of knowledge has been gained [3]. The discovery by Gaitan [4] that it is possible to drive a single stable bubble in a regime, where it emits light pulses of picosecond duration [5,6], called single bubble sonoluminescence (SBSL), has been encouraging scientists to explore the phenomenon and the associated effects with a multitude of experiments, theories and simulations. The experimental results show picosecond synchronicity [7], quasiperiodic and chaotic variability of inter-pulse times [8,9], a black body spectrum [10] and mass transport stability [11]. The theories to explain the source of SBSL range from hot spot, bremsstrahlung [12], collision induced radiation [13], and corona discharges [14] to non-classical light [15]. Numerical simulations have been focusing on the bubble dynamics, behavior of the gas content [12,16], properties in magnetic fields [17] and the stability of the bubble [18]. However, the final answer concerning the nature of SBSL still remains open.

The amount of energy concentration from low energy acoustic sound waves to 3 eV photons [5,19] raises the question, whether the effect can be up-scaled. In this paper, we report on experimental enhancement of SBSL

light production by a bimodal excitation of the bubble oscillation. The experiment follows an idea stated in [20]. We also present numerical simulations of multimode sound driving that reveal how multiharmonic excitation can adapt to the highly nonlinear bubble oscillation in the sense of a strong collapse.

The experimental setup is as follows: An air bubble is trapped in a water filled cell consisting of two piezoceramic cylinders connected via a glass tube [21]. The levitation cell (“Crum cell”) [22] is standing upright with a glass plate covering the lower end of the cell. The upper end remains open. A video camera pointing from the side allows for online monitoring of the experiment. The experiments were done with distilled and degassed water at room temperature and an ambient pressure of 1 atm. The bimodal driving signal $P_e(t) = P_1 \cos(2\pi ft) + P_2 \cos(2\pi 2ft + \phi)$ is produced by synchronized sine wave generators that allow to fix the amplitudes P_1 , P_2 , and the relative phase ϕ .

Using a multifrequency driving signal however is complicated by two facts. First, the transducers have a complex transfer function; second, the standing wave conditions at each frequency in the cell have to be obeyed [23]. Therefore, multifrequency driving results in space dependent phases and amplitude relations and thus in an effective sound signal $P_a(r, z, t)$ (with cylinder coordinates r, z of the levitation cell). To measure the amplitudes and relative phase that actually appear at the bubble position, a small hydrophone is used. The correct position is adjusted by first focusing the camera on the bubble and then inserting the hydrophone at the bubble site. The driving signal is digitally recorded and phases and amplitudes are recovered via a Fourier transform. The light flashes emitted at collapse are measured with a photomultiplier.

Levitating small oscillating bubbles of volume $V(t)$ in nonzero gravity is possible through the interaction with the driving sound field $P_a(r, z, t)$ which depends on space and time. The time averaged primary Bjerknes force [24]

$$F_B = -\langle V(t) \nabla P_a(r, z, t) \rangle \quad (1)$$

can overcome the buoyancy force and attract the bubble to a fixed position in space. Weakly sinusoidally driven bubbles of equilibrium radius R_0 are trapped near a pressure antinode if they are driven below their linear resonance (Minnaert) frequency $f_M = (2\pi R_0)^{-1} \sqrt{3\kappa p_0 / \rho} \approx 3/R_0$ [Hz] for the experimental conditions used here (with

polytropic exponent κ , ambient pressure p_0 , and liquid density ρ) [25]. However, the situation is more complicated for strongly driven bubbles [26] and also for multimodal excitation, where the standing wave pattern in the resonator, the Bjerknes forces and thus the bubble position are changed by a variation of the sound signal parameters P_1 , P_2 , and ϕ . The bubble oscillation responds to the sound signal at the trapping site.

In the experiment, we proceeded in the following way: For fixed drive amplitudes P_1 and P_2 , a bubble is injected into the fluid with a syringe. Once the bubble fixes itself spatially at a stable position, where the Bjerknes force equals the buoyancy force, the phase difference between the locked sine wave generators, one operating at $f = 23.4$ kHz and the other at $2f$, is sequentially increased while the sonoluminescence (SL) intensity and the bubble itself are monitored. Fig. 1 (lower) shows the SL intensity as a function of the phase difference for $P_1 \approx 1.25$ bars, $P_2 \approx 0.3$ bar. With increased phase difference two maxima appear in the light intensity. The dashed line indicates the maximum achievable SL intensity using single-mode driving. This value is obtained shortly before and after the two-mode experiment to allow direct comparison by keeping all other experimental conditions unchanged. It is seen that the two-mode driving yields 100% more SL intensity than the maximal single-mode driving. By further increase of P_1 and P_2 at selected phases an intensity gain of 300% can be achieved, as shown by the open circles. Beyond that, the bubble gets destroyed.

Fig. 1 (upper) reveals, that with increased phase difference the bubble traverses vertically through stable and (surface) unstable regimes.

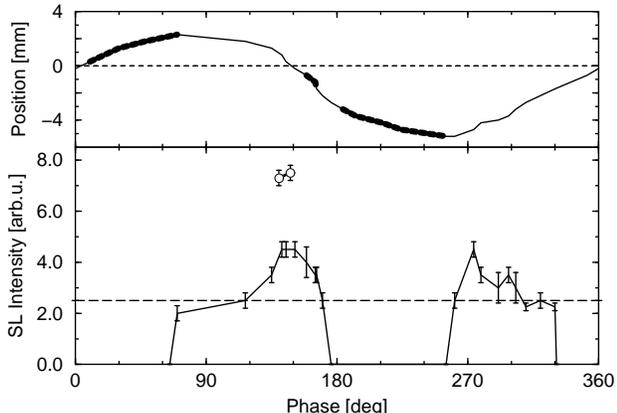


FIG. 1. Bubble response for two-mode driving as a function of the phase difference (in degrees) between the driving sinusoidal signal and its second harmonic. Upper: Vertical position of the bubble. The thick dotted lines denote unstable bubble behaviour. Lower: Photo current. The open circles show the maximum SL intensity achieved. The dashed line is the maximal photo current for pure sine wave driving.

Numerical simulations have been carried out using the Gilmore model [27] which describes the radial motion of a single bubble. The model includes the usual components of the Rayleigh-Plesset equation [28] like surface tension σ and liquid viscosity μ , and also the compressibility of the liquid to allow damping of the bubble motion by the shedding of shock waves.

$$\begin{aligned} \left(1 - \frac{\dot{R}}{C}\right) R\ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3C}\right) \dot{R}^2 \\ = \left(1 + \frac{\dot{R}}{C}\right) H + \left(1 - \frac{\dot{R}}{C}\right) \frac{R}{C} \frac{dH}{dt}, \quad (2) \\ H = \int_{p_\infty}^{p(R)} \frac{dp}{\rho}, \quad \frac{p(R, \dot{R}) + B}{p_0 + B} = \left\{ \frac{\rho}{\rho_0} \right\}^n, \\ c|_{r=R} = C = \sqrt{\frac{dp}{d\rho}} \Big|_{r=R} = \sqrt{c_0^2 + (n-1)H}, \\ p(R, \dot{R}) = \left(p_0 + \frac{2\sigma}{R_0}\right) \left(\frac{R_0^3 - a^3}{R^3 - a^3}\right)^\kappa - \frac{2\sigma}{R} - \frac{4\mu}{R} \dot{R}. \end{aligned}$$

R is the bubble radius, $R_0 = 5 \mu\text{m}$ its equilibrium radius, and C , ρ , and p are the speed of sound in the liquid, its density, and the pressure at the bubble wall, respectively. H is the enthalpy of the liquid. Parameters were set to $c_0=1500$ m/s, $\rho_0=998$ kg/m³, $p_0=1$ bar, $\kappa=4/3$, $\sigma=0.0725$ N/m, $\mu=0.001$ Ns/m³, $n=7$, $B=3000$ bars. $a=R_0/8.54$ is a hard-core van der Waals-term [28]. The pressure at infinity includes the multimodal driving pressure: $p_\infty = p_0 + P_e(t)$, $P_e(t) = \sum_{m=1}^M P_m \cos(2\pi m f t + \phi_m)$.

First we calculated the driving sound signal that would lead to the most violent collapse, indicated by the smallest minimum radius during a bubble oscillation cycle using the above equation. The search for suitable pressures P_m and phases ϕ_m was carried out by a heuristical optimization algorithm [29] with the boundary condition of a constant driving signal power, i.e., $P_e^2 = \sum_{m=1}^M P_m^2 = \text{const}$. P_e was fixed to 1.3 bars and the driving frequency was the same as in the experiment. Comparing equal power signals is convenient, because the power stays constant upon phase changes, making it possible to compare numerics and experiments.

Fig. 2 shows the driving pressure and the bubble response of different driving signals. A strong increase in the maximum radius can be seen already by adding just the second harmonic to a sine wave. The numerically computed optimal phase difference is 166.4 deg and the individual amplitudes are $P_1 = 1.026$ bars and $P_2 = 0.798$ bar. The radius and the adiabatically calculated temperature around the collapses are shown in Fig. 3. It is seen, that the bubble radius at collapse is decreased by a large amount and is approaching the van der Waals hard core already for the two-mode driving.

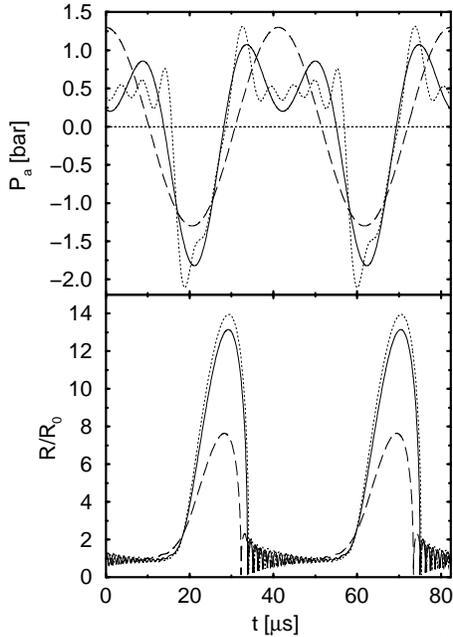


FIG. 2. Time series of the driving (top) and the radius of calculated bubble collapses (bottom) for single (dashed) and optimized multimode driving signals (two-mode: line, eight-mode: dotted).

Also the maximum temperatures almost double. The higher-mode driving signals are better adapted to the nonlinear bubble oscillation than the sine signal: They show a deeper rarefaction phase before collapse, followed by a more rapid rise to the compression phase during collapse.

The calculations for optimal eight-mode driving exhibit only small additional gain compared to bimodal driving. Because of the increased difficulties regarding the spatial stability of bubbles in the resulting complicated sound field, an eight-mode driving may not be worth being considered experimentally. Though two-mode driving is an early truncation of a series expansion, one sees that already this approximation shows a trend for a more intense driving of this nonlinear system.

The optimal results are located on a single broad plateau in parameter space. This is in contrast to the experimental finding of two maxima. To understand the reason of this obvious discrepancy, the bubble model [Eq. (2)] has been integrated numerically along with the primary Bjerknes [Eq. (1)] and the buoyancy forces to examine spatial dependencies. This is also motivated by the observation that the vertical position of the bubble is altered when the phase is changed (Fig. 1 upper). The change of position leads to different effective excitation amplitudes for f and $2f$ and thus to a more complex scenario. The system of equations is integrated using the spatially dependent driving force

$$P_e(t) = P_1 \cos(2\pi ft) \cos(k/2 z) + P_2 \cos(2\pi 2ft + \phi) \cos(2kz - \pi/2)$$

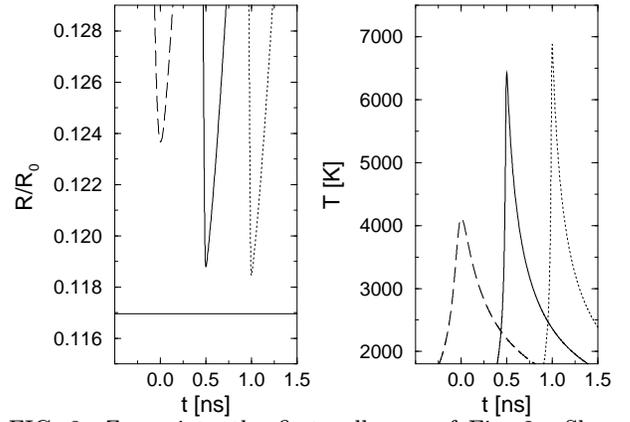


FIG. 3. Zoom into the first collapses of Fig. 2. Shown are the curves for single-mode (dashed), optimized two-mode (line) and optimized eight-mode (dotted) driving. The time series of the bubble collapses exhibit a decrease in minimum radius (left) and increase in the adiabatically calculated temperature (right) as a function of the number of modes in the driving sound. The minimum radius comes very close to the van der Waals hard core, shown by the horizontal line in the left graph. The time axis is shifted so that the collapses take place at 0, 0.5, and 1 ns, respectively.

(k is the acoustic wavenumber $2\pi f/c_0$, $P_1 = 1.25$ bars, $P_2 = 0.357$ bar, $f = 23.4$ kHz). The spatial modes are approximately the same as the experimental modes, which have been measured with a needle hydrophone. The points in vertical z -space where the Bjerknes force vanishes and the stability criterion is met represent the position of the bubble. The resulting minimum radii are shown in Fig. 4. Comparing this with the experimental results in Fig. 1 shows a very close agreement.

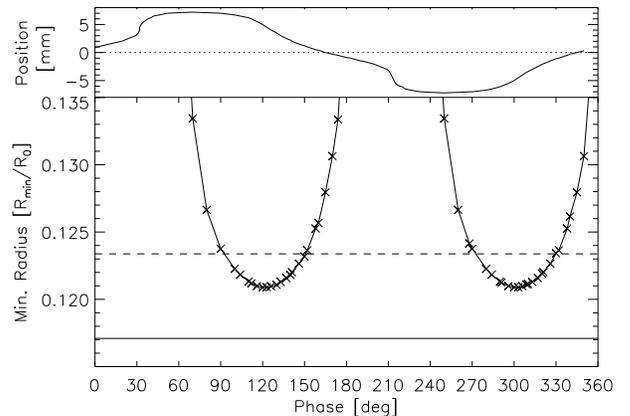


FIG. 4. Numerically calculated vertical bubble position (upper) and resulting minimum radius (lower) as a function of the phase difference for double harmonic driving of a bubble. The dashed line in the lower plot is the minimum radius for single frequency driving with the same power. The solid straight line is the van der Waals hard core.

The almost sinusoidal variation of the position of the bubble gives rise to two minima of the minimal radius/phase dependence. Each of these minima is smaller

than the one of the single-mode driving. The minima coincide with the experimental observation of increased SL intensity. The slight asymmetry in the experiment can be described by the difference in acoustic impedance of the glass bottom and the open top of the cell. Also, imperfect standing waves may lead to small traveling components in the experimental driving. Changing the amplitude ratio of the driving signal closer to unity while keeping the power constant results in a complex scenario of stable bubble positions and effective drivings including hysteretic jumps.

In summary, we have shown that a bimodal sound excitation can enhance light production of SBSL. Though spatial modes play a crucial role in double harmonic driving, it increased the photo current to a gain of maximally 300% compared to sine excitation. We suppose that multifrequency driving can shift the bubble oscillation to a regime of strong stable SBSL which is not reachable by pure harmonic driving. Numerical simulations of an acoustically driven bubble including Bjerknes and buoyancy forces show that the increased SBSL light intensity is caused by a larger compression. To give quantitative estimates, however, elaborate models have to be considered that include gas dynamic equations for the interior of the bubble and thus can model the shedding of a shock wave inside a bubble [12,16,30].

Other methods have been proposed to increase the violence of bubble collapses. For example, calculations for thermonuclear D-D fusion in D₂O within this context have been done using a large pressure pulse superimposed on a sine wave [16]. However, whether advanced forcing by higher modes is large enough to achieve a reasonable neutron production rate is an open question. Apart from sonoluminescence, the increase of cavitation strength by means of optimized multiharmonic sound signals [20] can also be of use in the context of sonochemistry [31] and related areas, where higher reaction rates could be induced.

The authors wish to thank R. G. Holt and W. Lauterborn for stimulating discussions and the TU Darmstadt for making the research possible. The work has been funded through the SFB 185 "Nichtlineare Dynamik" of the DFG.

[1] W. Lauterborn, and J. Holzfuss, *J. Bif. and Chaos* **1**, 13 (1991); W. Lauterborn, and J. Holzfuss, *Phys. Lett. A* **115**, 369 (1986).
 [2] N. Marinenco and J. J. Trillat, *Proc. R. Acad. Sci.* **196**, 858 (1933).
 [3] A. J. Walton and G. T. Reynolds, *Adv. Phys.* **33**, 595 (1984); F. R. Young, *Cavitation* (McGraw-Hill, London, 1989).
 [4] D. F. Gaitan, L. A. Crum, C. C. Church, and R. A. Roy, *J. Acoust. Soc. Am.* **91**, 3166 (1992).
 [5] B. P. Barber, S. J. Putterman, *Nature* **352**, 318 (1991).

[6] B. Gompf, R. Günther, G. Nick, R. Pecha, and W. Eisenmenger, *Phys. Rev. Lett.* **79**, 1405 (1997).
 [7] B. P. Barber, R. Hiller, K. Arisaka, H. Fetterman, and S. Putterman, *J. Acoust. Soc. Am.* **91**, 3061 (1992).
 [8] R. G. Holt, D. F. Gaitan, A. A. Atchley, and J. Holzfuss, *Phys. Rev. Lett.* **72**, 1376 (1994).
 [9] J. Holzfuss, R. G. Holt, D. F. Gaitan, and A. A. Atchley, in: *Fortschritte der Akustik - DAGA 93*, (DPG GmbH, Bad Honnef, 1993), pp. 329-332.
 [10] R. Hiller, S. P. Putterman and B. P. Barber, *Phys. Rev. Lett.* **69**, 1182 (1992).
 [11] R. G. Holt and D. F. Gaitan, *Phys. Rev. Lett.* **77**, 3791 (1996).
 [12] C. C. Wu and P. H. Roberts, *Phys. Rev. Lett.* **70**, 3424 (1993).
 [13] L. Fromhold, *Phys. Rev. Lett.* **73**, 2883 (1994).
 [14] T. Lepoint, N. Voglet, L. Faille, and F. Mullie, in: *Bubble Dynamics and Interface Phenomena*, edited by: J. R. Blake, J. M. Boulton-Stone, and N. H. Thomas, (Kluwer Academic Publishers, Dordrecht, 1994), pp. 321-333.
 [15] J. Schwinger, *Proc. Natl. Acad. Sci. USA* **89**, 4091 (1992); C. Eberlein, *Phys. Rev. Lett.* **76**, 3842 (1996).
 [16] W. C. Moss, D. B. Clarke, J. W. White, and D. A. Young, *Phys. Lett. A* **211**, 69 (1996).
 [17] T. Chou and E.G. Blackman, *Phys. Rev. Lett.* **76**, 1549 (1996).
 [18] M. P. Brenner, D. Lohse, D. Oxtoby, and T. F. Dupont, *Phys. Rev. Lett.* **76**, 1158 (1996); M. P. Brenner, D. Lohse, and T. F. Dupont, *Phys. Rev. Lett.* **75**, 954 (1995).
 [19] B. P. Barber, R. A. Hiller, R. Löffstedt, S. J. Putterman, and K. R. Weninger, *Phys. Rep.* **281**, 65 (1997).
 [20] R. Mettin, J. Holzfuss, and W. Lauterborn, in: *Fortschritte der Akustik - DAGA 95*, (DPG GmbH, Bad Honnef, 1995), pp. 1147-1150.
 [21] R. G. Holt, J. Holzfuss, A. Judt, A. Phillip, and S. Horsburgh, in: *Frontiers of Nonlinear Acoustics: Proceedings of the 12th ISNA*, edited by: M.F. Hamilton and D.T. Blackstock, (Elsevier Science Publishers Ltd., London, 1990), pp. 497-502.
 [22] L. A. Crum, *J. Acoust. Soc. Am.* **68**, 203 (1980).
 [23] The largest resonances, Q-factors and amplitudes relative to the main are: (23.4 kHz, 84, 1), (44.4, 21, 0.12), (45.8 kHz, 89, 0.19), (46.8 kHz, 67, 0.17).
 [24] L. A. Crum, *J. Acoust. Soc. Am.* **57**, 1363 (1975).
 [25] M. Minnaert, *Phil. Mag.* **16**, 235 (1933).
 [26] I. Achatov, R. Mettin, C.D. Ohl, U. Parlitz, and W. Lauterborn, *Phys. Rev. E* **55**, 3747 (1997).
 [27] F. R. Gilmore, California Institute of Technology Report No. 26-4, (1952).
 [28] R. Löffstedt, B. P. Barber and S. J. Putterman, *Phys. Fluids A*, **5** 2911 (1993).
 [29] We used the algorithm `amebsa` from: W.H. Press, S.A. Teukolsky, W.T. Vetterling, and B.R. Flannery, *Numerical Recipes in C* (Cambridge University Press, Cambridge 1992) 2nd ed.
 [30] W. C. Moss, D. B. Clarke, and D. A. Young, *Science* **276**, 1398 (1997).
 [31] *Ultrasound: Its Chemical, Physical and Biological Effects* edited by K. S. Suslick, (VCH, New York, 1988)