Sonoluminescence: When bubbles glow

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In single-bubble sonoluminescence, an ultrasound-driven microbubble undergoes violent oscillations and converts part of the energy of the sound field into visible light. Since its discovery ten years ago, a consistent theory of the dynamics and stability of the bubble has been developed. The approach includes the dissociation of molecular gases inside the collapsing bubble to explain striking differences in experiments with various molecular and noble gases. A natural extension of the theory incorporates the light emission itself, identifying its mechanism as thermal bremsstrahlung from the optically thin (transparent) bubble. A direct and systematic comparison of the results of the theory with experiment shows good agreement in all characteristic features.

WHEN Felipe Gaitan was working on his Ph D at the University of Mississippi in 1990, he hoped for new insights into the dynamics of a single, ultrasonically driven bubble. He was thrilled to discover that he could make the bubble emit a steady glow of bluish light. This phenomenon is now known as (single bubble) *sonoluminescence* (SL)[1–3]. Converting sound into light is not only a fascinating feat, but is remarkable for the physicist as well: the energy density in the emitted photons is about 12 orders of magnitude greater than that in the driving sound field!

An additional appeal comes from the simplicity of the experimental set-up (Figure 1), whose main component is a flask filled with partially degassed water (the gas concentration is smaller than the saturation concentration). A single bubble is injected into the flask; a standing ultrasound wave (with typical frequencies of 20–40 kHz and pressure amplitudes of 1.2–1.5 atm) traps the bubble at the centre of the vessel and at the same time excites it to highly nonlinear radial oscillations. The bubble collapses very violently once per driving cycle and concentrates the energy sufficiently to allow for the emission of a very brief pulse of light. Even though the bubble is only a few micrometers across, its light is visible to the naked eye.

SL immediately became the target of the efforts of many research groups, but several years after Gaitan's discovery the mechanism of light emission was still unknown, and a number of other unresolved puzzles had appeared. These included the extreme stability of the phenomenon (over hours or days) in a very narrow range of driving pressures or the increase of the light intensity with decreasing water temperature. A series of experiments with different gas mixtures revealed perhaps the most astonishing result: a tiny amount of a *noble gas* (like 1% argon contained in air) is necessary for the above-mentioned stability – without it the bubbles do not live longer than a couple of minutes.

Almost all of these open questions have been answered satisfactorily in the past few years, and the increased understanding of SL bubbles has finally led to a plausible model for the light emission, too. The starting point for the work in our group was the stability of the bubbles, for which ground-breaking work was done by Andrea Prosperetti[4–6] and also by many others[7–11]

Figure 1. Photograph showing a water-filled flask used in the single-bubble SL experiment, with a glowing bubble at its center. The annular object on the flask wall is one of the two piezoelectric transducers which generate the sound field.

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It soon became clear that the bubble can only be stable if (i) perturbations of its spherical shape stay small[12], and (ii) the gas content of the bubble (its mass) stays constant in time and is not changed through diffusive mass exchange with the dissolved gas in the water[13]. The validity or violation of both criteria can be checked directly from the *dynamics* of the bubble oscillation. As a result, the shape stability condition (i) gives upper bounds for bubble mass and driving pressure: if the bubble contains too much gas or if it is driven too strongly, it becomes shape unstable and finally fragments into many microbubbles. The diffusive stability condition (ii) singles out one specific gas content for a bubble in stable diffusive equilibrium with its surroundings (at given gas concentration and driving pressure). Obviously, both conditions (i) and (ii) can be fulfilled simultaneously only in a certain domain of parameter space. Calculating this region[13] gives excellent agreement with experiments that use *pure argon* or other pure noble gases, where very strong degassing to about 0.2–0.4% of saturation is necessary for stable bubbles.

Until recently, however, there was a huge discrepancy between the theoretical predictions for air (which are almost identical to those for pure noble gases) and the observed stability of SL bubbles at 20–40% of saturation for *air*, i.e. for about 100 times greater concentration of dissolved gas. We found a very simple rationalization for this phenomenon[14]: at the high temperatures inside the collapsing bubble all molecular gases (oxygen, nitrogen) in the air *dissociate*, and the resulting radicals form products that dissolve in the surrounding water. The only constituent of air which cannot dissociate is argon, and it is all that remains of the air bubble. As about one part in 100 of air is argon, the actual relevant concentration of dissolved gas is 100-fold smaller than the total air concentration, and is thus again in outstanding agreement with theory. These predictions have meanwhile been confirmed by quantitative experiments conducted by Robert Apfel and his group at Yale[15]. This 'burning off' of molecular gases also explains why a small amount of noble gas is necessary to ensure stable SL.

As material parameters are changed, the boundaries for bubble stability shift, too. This offers a natural explanation for the increase in light intensity upon cooling of the water[16]: the stability diagrams show that the largest stable bubbles can also be driven at the highest pressure amplitudes. Therefore, they show the most violent collapses and emit the most intense light. When the water is cooled, the parameter regime of stable SL widens because (among other effects) the viscosity of water and the solubility of the gas increase. Thus, larger bubbles can be stable at higher drivings and emit light pulses of considerably higher intensity. Similar effects might be achieved by using non-aqueous fluids instead of water, exploiting their different viscosities, surface tensions, etc.

The picture of light emission itself, the spectacular feature of sonoluminescing bubbles, has also cleared up a lot recently. Bruno Gompf, Wolfgang Eisenmenger and their collaborators at the University of Stuttgart[17,18] obtained the first time-resolved measurement of the light pulse, using single-photon correlation. Depending on gas concentration and driving pressure amplitude, the temporal width of the pulse was found to be 60–300 ps. These results improved and corrected earlier estimates of US-based groups[3,19] which suggested much smaller pulse widths. Another important result obtained at Stuttgart is the near independence of the pulse width on the wavelength of the light (the pulse is as long in the red part of the spectrum as in the ultraviolet). The observations contradict the most simple model one could postulate for SL light emission: if SL was thermal black body radiation from the hot bubble, the pulse widths should be larger still, and should vary with wavelength by as much as a factor of two.

It turned out[20], though, that the black body model needs but a small modification, which follows directly from the physical properties of the gas in the bubble: to emit black body radiation, the bubble has to be *black*, i.e. an ideal absorber for all wavelengths. The calculated mean free path of photons in the gas of the collapsed bubble, however, is always larger than the bubble radius, so that the bubble is *transparent* for all the photons it emits. Thus, our picture of SL light emission is the following[21]: (1) The violent collapse of the bubble heats the gas to about 20,000 K (just like the air in a bicycle tyre, only at a much more extreme compression). (2) At these temperatures a small fraction of noble gas atoms are *ionized* and release free electrons into the gas. (3) Bremsstrahlung of the free electrons in the field of the ions and neutral atoms as well as radiative recombination are sources of photon emission in the weakly ionized gas. The photons are not reabsorbed in the transparent gas, and can be observed directly for wavelengths > 200 nm (smaller wavelengths are absorbed in the surrounding water). (4) After about 100 ps the temperature in the bubble begins to fall again. The ionization shows an extremely sensitive dependence on temperature, and thus drops precipitously. Therefore, photon emission at *all* wavelengths ceases simultaneously.

This approach explains both the typical width of SL pulses and their equal length in the red and UV spectral regimes. The spectrum of emitted photons in this case carries information about the light emitting processes, unlike a pure black body radiation, which has a universal spectrum independent of the microscopic emission processes. Our model[21], which does not contain any adjustable parameters, is indeed able to reproduce not only the observed photon numbers and widths of SL pulses, but also their spectra satisfactorily.

All above-mentioned calculations are computationally relatively inexpensive, just as those for bubble shape stability and diffusive stability. Therefore, we can obtain results for many parameter combinations in the parameter space of SL, and can for the first time compare the results directly to whole sets of reliable experimental data, see

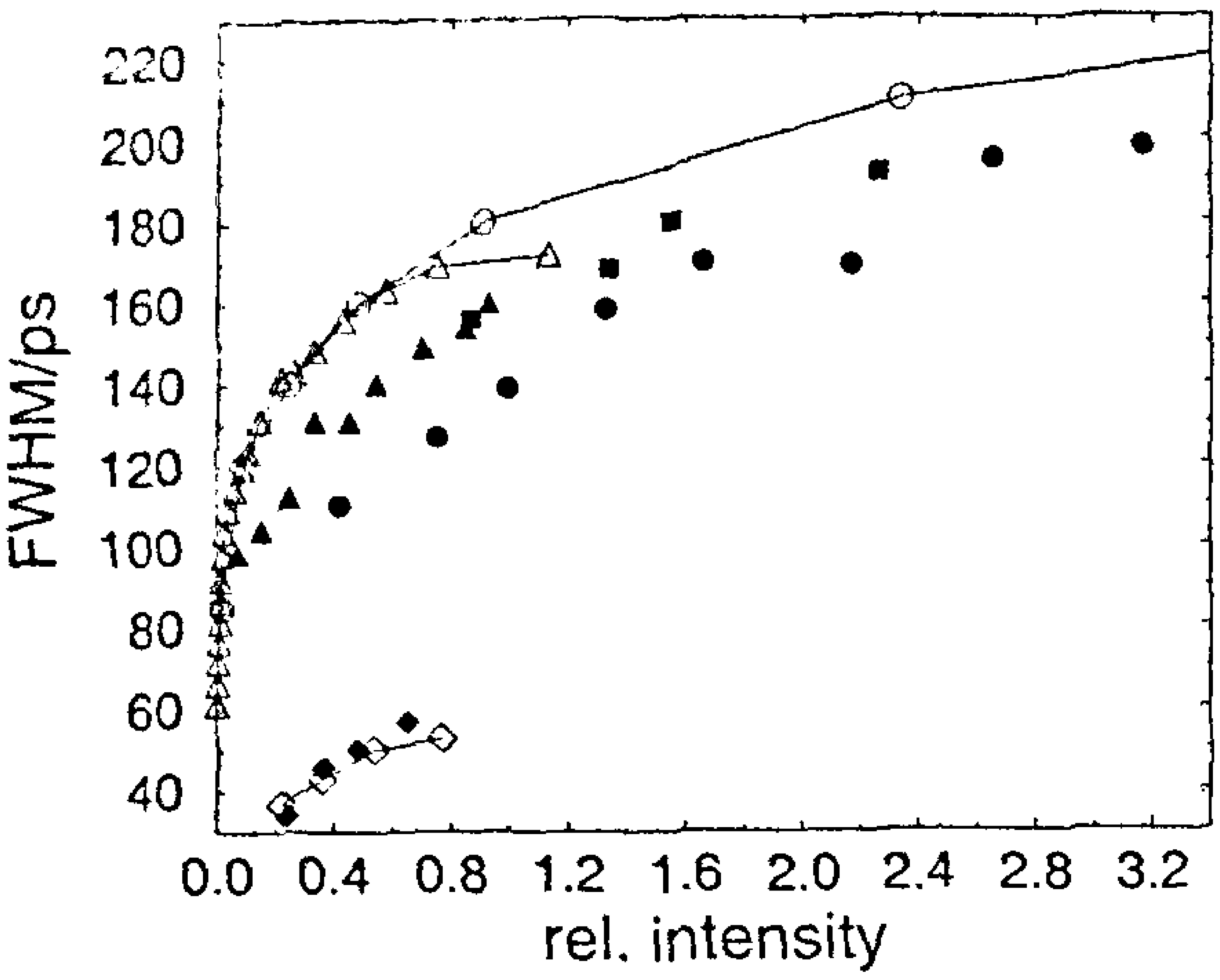

Figure 2. Comparison of measured SL pulse widths and intensities (filled symbols) and predictions from the theory outlined here (open symbols). Data are for xenon (circles) and argon (triangles) at a gas concentration of 0.4% saturation, and for air (diamonds) at 3%, for which the theoretical calculations used argon at 0.03% because of the 'burning off' of oxygen and nitrogen in air. The driving frequency is 34 kHz in all cases.

Figure 2. Here we show the width of the SL light pulse as a function of its intensity, computed theoretically and compared to various experimental results for different concentrations of dissolved argon, xenon, and air. The gas concentration is the same for all points in one data set, while the driving pressure is varied. The longest and most intense pulses represent the largest drivings. The theoretical predictions of our model, based only on experimental parameters and material constants (no fit parameters!) agree well with the data, capturing the qualitative trend in all cases and the quantitative value for most data points[21,22].

Note that a comparison like this tests all the salient features of the model: to calculate the light emission, one has to know the gas content of the bubble that is stable at a given driving pressure, necessitating a bubble stability computation. The dissolved gas concentration has to be modified for air as only argon remains in the bubble – for the case in Figure 2, where the experiment used air degassed to 3% of saturation the computation must be done for argon at 0.03%. Finally, the mutual dependence of pulse widths and intensities probes our physical concept of SL light emission. The good agreement with experiment provides strong support for the validity of our basic assumptions. We stress that our theoretical description makes use of nothing but well-known physical principles. SL is, without doubt, one of the most fascinating phenomena of physics in the past decade – its main features could now be explained using quite old-fashioned physics.

While the theoretical concept of SL becomes more and more complete, practical applications for the newly-acquired knowledge begin to emerge. With the dissociation and ionization reactions in its interior, a SL bubble is a micro-laboratory for high temperature chemistry. Perhaps it will be possible to control the reaction by adjusting external parameters like driving pressure or water temperature. Ultrasonically-driven clouds of many bubbles are already in use as 'catalysts' for chemical reactions (sonochemistry)[23,24].

Medicine offers another application for micrometer-sized bubbles: In ultrasound diagnostics, they are ideal tracers or contrast agents because of their huge scattering cross-sections for diagnostic ultrasound[25,26]. With our present understanding of bubble dynamics, largely due to SL research, a deliberate 'design' of contrast agents is now possible.

Without Gaitan's initial experiments, the perspectives in the above-mentioned applications would probably be less bright. SL, with its multitude of phenomena from diverse fields of physics, engineering, and chemistry, thus highlights the importance of fundamental research as well as the necessity for interaction between the disciplines.

1. Gaitan, D. F., Ph D thesis, University of Mississippi, 1990.
2. Crum, L. A., *Phys. Today*, 1994, **47**, 22.
3. Barber, B. P. *et al.*, *Phys. Rep.*, 1997, **281**, 65.
4. Prosperetti, A., *Q. Appl. Math.*, 1977, **34**, 339.
5. Plesset, M. and Prosperetti, A., *Annu. Rev. Fluid Mech.*, 1977, **9**, 145.
6. Prosperetti, A., *J. Fluid Mech.*, 1991, **222**, 587.
7. Eller, A., *J. Acoust. Soc. Am.*, 1969, **46**, 1246.
8. Eller, A. and Crum, L., *J. Acoust. Soc. Am. Suppl.*, 1970, **47**, 762.
9. Fyrillas, M. M. and Szeri, A. J., *J. Fluid Mech.*, 1994, **277**, 381.
10. Brennen, C. E., *Cavitation and Bubble Dynamics*, Oxford University Press, Oxford, 1995.
11. Leighton, T. G., *The Acoustic Bubble*, Academic Press, London, 1996.
12. Brenner, M., Lohse, D. and Dupont, T., *Phys. Rev. Lett.*, 1995, **75**, 954.
13. Hilgenfeldt, S., Lohse, D. and Brenner, M. P., *Phys. Fluids*, 1996, **8**, 2808.
14. Lohse, D. *et al.*, *Phys. Rev. Lett.*, 1997, **78**, 1359.
15. Ketterling, J. A. and Apfel, R. E., *Phys. Rev. Lett.*, 1998, **81**, 4991.
16. Hilgenfeldt, S., Lohse, D. and Moss, W., *Phys. Rev. Lett.*, 1998, **80**, 1332.
17. Gompf, B. *et al.*, *Phys. Rev. Lett.*, 1997, **79**, 1405.
18. Pecha, R., Gompf, B., Nick, G. and Eisenmenger, W., *Phys. Rev. Lett.*, 1998, **81**, 717.
19. Moran, M. J. *et al.*, *Phys. Res.*, 1995, **B96**, 651.
20. Moss, W., Clarke, D. and Young, D., *Science*, 1997, **276**, 1398.
21. Hilgenfeldt, S., Grossmann, S. and Lohse, D., *Nature*, 1999, **398**, 402.
22. Hilgenfeldt, S., Grossmann, S. and Lohse, D., *Phys. Fluids*, 1999, **11**, 1318.
23. Suslick, K. S., *Science*, 1990, **247**, 1439.
24. Flint, E. B. and Suslick, K. S., *Science*, 1990, **253**, 1397.
25. See the articles in *Advances in Echo Imaging Using Contrast Enhancement* (eds Nanda, N. C. and Schlief, R.), Kluwer Academic Publishers, Dordrecht, 1993.
26. Hilgenfeldt, S., Lohse, D. and Zomack, M., *Eur. Phys. J.*, 1998, **B4**, 247.

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