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Sonoluminescence

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Pulsed light scattering off of a gas bubble trapped in a fluid by a strong sound field indicates that 'sonoluminescence' is accompannied by a high Mach number implosion of the bubble's wall. Although these measurements support the imploding shock wave model of sonoluminescence there remain many unknowns including the effects of impurities, fluid temperature, noble gas doping as well as the bubble size and light-emitting mechanism.

1. Introduction

As the intensity of a standing sound wave is increased the pulsations of a bubble of gas located at a velocity node become sufficiently nonlinear so as to emit ultraviolet flashes of light (Hiller *et al.* 1992*a*, 1994) with a duration measured in picoseconds (Barber & Putterman 1991). The acoustic resonator can be tuned so that the flashes of light come out with a clocklike synchronicity (Barber & Putterman 1991; Barber *et al.* 1992) one flash for each cycle of sound with the jitter in the time between flashes also being measured in picoseconds. This phenomenon (sonoluminescence or 'SL') is interesting because it is the only means of generating picosecond flashes of light that does not use a laser. Also, the efficiency with which the frequency of the sound field (30 kHz) is upshifted by about eleven orders of magnitude (Barber *et al.* 1991) to make light (1000 THz) is remarkable. The amount of energy emitted as light is greater than the acoustic energy lost to viscous damping in the fluid in the absence of the bubble.

As yet water appears to be the friendliest fluid for SL. Many gases yield SL in water (Hiller *et al.* 1994) but with air bubbles it is essential that the water be degassed (Gaitan 1990; Gaitan *et al.* 1992). This distinguishes synchronous SL from the transient SL (Frenzel & Schultes 1934) which is emitted by clouds of cavitating bubbles and runs best at saturation. Pure diatomic gases give little or no light so that the 1% Argon content of air has a dramatic effect (Hiller *et al.* 1994). Sonoluminescence can be seen in other liquids (Weninger *et al.* 1995) but in these cases its observation is greatly enhanced by the use of pure Xenon gas bubbles. In virtually all cases cooling the host fluid greatly increases the light output (Hiller *et al.* 1992; Weninger *et al.* 1995).

2. Bubble motion

Central to SL are the driven pulsations of a bubble of gas that is trapped in a fluid by a strong sound field. Figure 1 displays light scattering measurements of the

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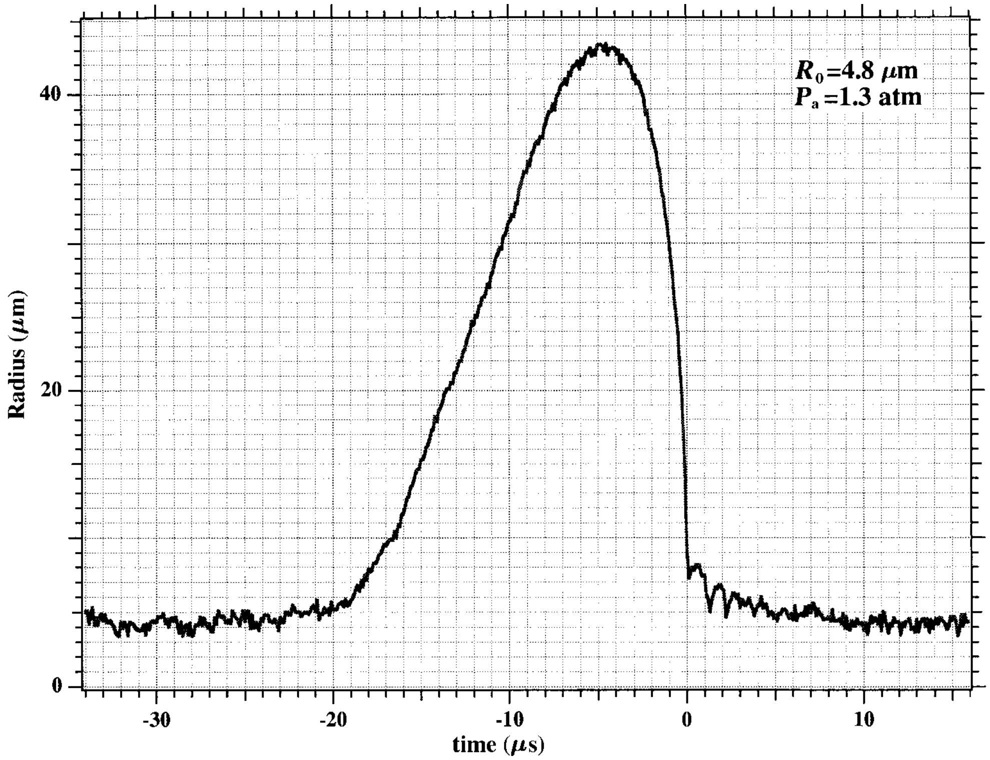

Figure 1. Bubble radius as a function of time for one cycle of the applied sound field. The ambient radius R_0 is 4.8 μm and the imposed sound field is 1.3 atm. Measurements taken for air dissolved into water at a partial pressure of 150 mm and driven at a frequency of 22.9 kHz.

bubble motion. During the rarefaction part of the acoustic cycle when the net pressure goes negative the bubble expands. After the bubble reaches its maximum size it undergoes a runaway collapse (Rayleigh 1917) which is then followed by bounces whose period is determined by the pulsating resonances of a free (i.e. undriven) bubble in a fluid. Figure 2 shows a detail of this free ringing. Figure 3 shows a highly detailed measurement of the bubble motion as its radius approaches its minimum value at the bottom of the Rayleigh collapse. The slope of this curve reaches over four times the speed of sound in the ambient gas. The motion has become supersonic and the fluid motion no longer obeys the low mach number criterion that was imposed by Rayleigh in order to derive his equation for the bubble's motion from the Euler hydrodynamics. It has been proposed (Barber & Putterman 1992; Greenspan & Nadim 1993; Löfstedt 1992, 1993; Wu & Roberts 1993) that as a consequence of the fast collapse the bubble launches an imploding shock wave which now runs through the compressed gas increasing its strength and further concentrating the energy as it heads towards the origin. For the measurements shown in figure 3 the temporal resolution is 500 ps. Since the bubble turns around within one datapoint its acceleration at the minimum is over $3\times10^{11}g$ (!) and the acceleration of the shock front must be even greater. At this moment of great stress and energy focusing the flash of light is emitted; which effect now goes beyond the theoretical purview of the Euler/Navier–Stokes hydrodynamics.

These measurements constitute a pulsed version of the Mie scattering technique (Barber & Putterman 1992; Putterman 1995) that has been used to probe bubble

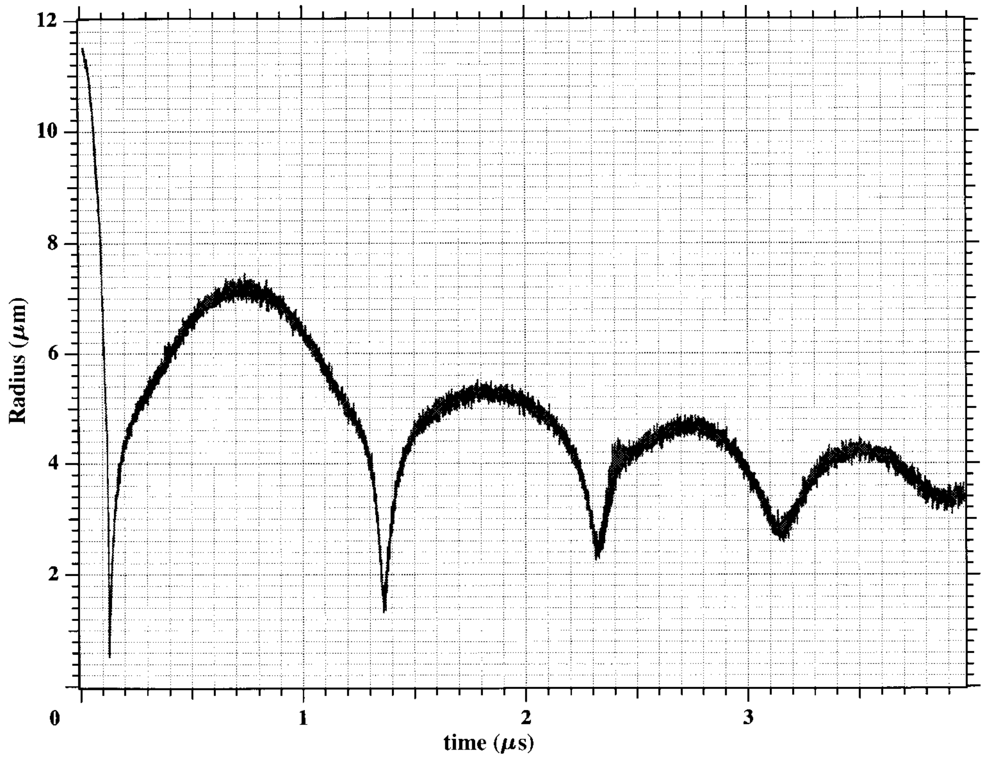

Figure 2. Detail of bubble radius as a function of time showing the free ringing motion that occurs after the Rayleigh collapse.

motion. If light scattered by the bubble is collected in a large enough solid angle (say 30° around 60° from the forward direction) then the bubble radius 'R' is proportional to the square root of the intensity of the collected light.

Figure 4 demonstrates a difficulty encountered when $R(t)$ is determined from the time dependence of the scattered light when the bubble is subjected to continuous illumination (Barber & Putterman 1992). The dashed line is a solution to the Rayleigh–Plesset equation of bubble motion with input parameters (ambient radius and acoustic amplitude) chosen so that this curve yields an approximate fit to the data shown in figure 3. The flash of SL is represented by a 'delta' function which meets $R(t)$ at its minimum. If the scattered light plus the flash are convolved with the detector response one obtains the solid line. This curve (Barber & Putterman 1992) is typical of the resolution obtainable with continuous illumination. In this case the finite response time of the photodetector averages together light scattered at different times thus hindering the temporal resolution of the motion. When the bubble is quickly collapsing the detector response function produces a rate of implosion that is slower than the actual motion as shown in the figure. In addition to the scattered laser light the flash of SL is also recorded by the detector. Its response time can also shift the timing of the minimum in $R(t)$ relative to the flash of SL as shown.

To beat the finite resolution time of the detector (and obtain the improved data displayed in figure 3) we scattered short (in this case 100 fs) pulses of light off the bubble at precisely determined times. In this way the integrated response of the detector can be attributed to these points in time. The flash of SL always determined

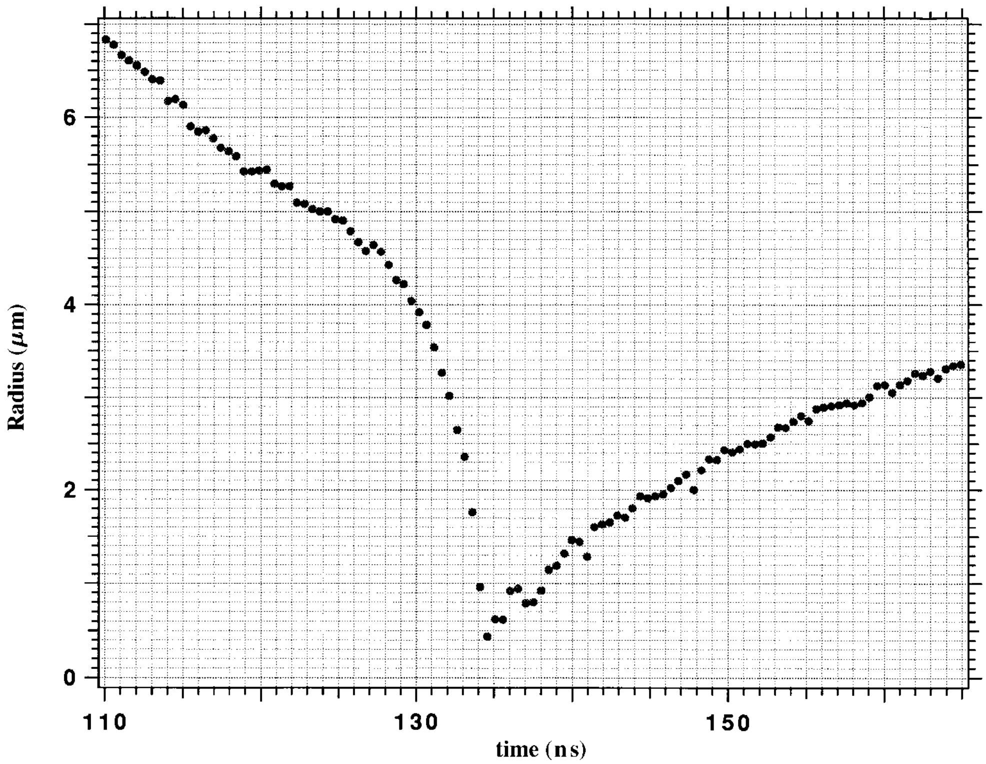

Figure 3. Nanosecond resolution of the bubble dynamics at the moment of collapse. The spacing between points is 500 ps. Data were acquired by scattering femtosecond flashes of 410 nm light off of the bubble at a repetition rate of 76 MHz. Each bin records the average of 88 measurements. The ray optics limit was used to calculate $R(t)$ from the collected scattered light. Deviations from this approximation lead to 10% increases in the plotted R between five and six μm and to a 10–15% increase in the slope between about one and two μm.

the reference time. Since the flash to flash jitter of SL is so low data accumulated over many cycles (as the probe to SL times were varied) could be combined to form figure 3.

The fast collapse of the bubble also launches a strong sound pulse of short duration out into the fluid . Shown in figure 5 is the response of a high bandwidth PVDF needle hydrophone (Precision Acoustics) placed 1 mm from the bubble. The resolution is limited by the 10 ns rise time of the microphone. If the measured peak dynamic pressure at a distance r from the bubble is $p_s(r)$ and if the shock is launched when the radius is R_c with a temporal width t_s then the actual amplitude of the *outgoing* shock at the time of launch is roughly given by $rp_s(r)(10\,\text{ns})/R_c t_s \Gamma(r)$, where Γ is the attenuation of the pulse due to viscous losses as it travels through the liquid to the microphone. For an ideal fluid $\Gamma = 1$, but for GHz waves in water Γ is already $1/e$ for $r = 100\,\mu\text{m}$. The overall deconvolution factor can therefore be very large. Its determination awaits improved methods of experimentation.

3. Conclusion and the unknowns of sonoluminescence

The figures indicate that SL is accompanied by the supersonic collapse of a pulsating bubble of gas that is trapped by a sound field in a fluid. The strongly supersonic

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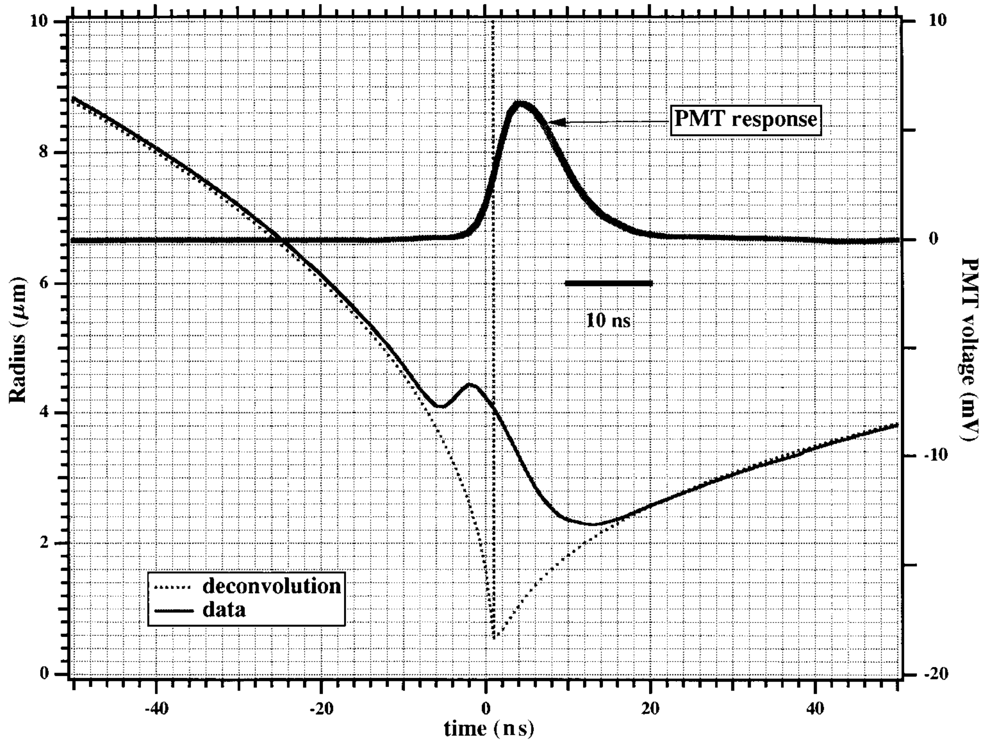

Figure 4. Effect of detector bandwidth on continuous wave light scattering. The dashed line is a fit of the measured $R(t)$, near the moment of collapse,to the Rayleigh–Plesset equation of bubble motion. Superimposed at the minimum radius is a 'delta' function peak for the flash of SL. When convolved with the detector response function the solid line is obtained. The effect of the detector response on the disparate time scales can lead to the mistaken impression that the flash is emitted before the minimum. The purpose of the pulsed measurements (figure 3) was to beat the limitations imposed by the detector response. The response of the photodetector (PMT) to a delta function of light is shown, with positive sign, in the upper trace.

motion suggests that the bubble launches an imploding shock wave. The main appeal of the shock wave model is that it provides a means of focusing energy by many orders of magnitude. The central success (Wu & Roberts 1993) is that it predicted that the bubble wall would reach a Mach number of about 4–5 before launching the shock. There is, however, no direct experimental observation of the imploding shock within the bubble.

While the shock wave model provides an enticing picture for the concentration of energy which leads to SL it nevertheless has a number of limitations. First it does not explain the range of sound field intensities for which SL can be observed (Barber *et al.* 1994). Furthermore this model does not explain the observed bubble radii which in many cases (e.g. air in water) cannot be accounted for by diffusive equilibrium (Barber *et al.* 1995; Löfstedt *et al.* 1993, 1995). The shock wave picture does not explain the role of noble gas dopants or the effects of cooling the fluid. The shock wave model provides no way to understand why water is the friendliest fluid for SL or why the spectra of bubbles in water and commercially available (Isotech) heavy water (Hiller & Putterman 1995) can be dramatically different.

Theoretical approaches to sonoluminescence generally separate the bubble dynamics from the process of light emission. The energy concentration brought about by

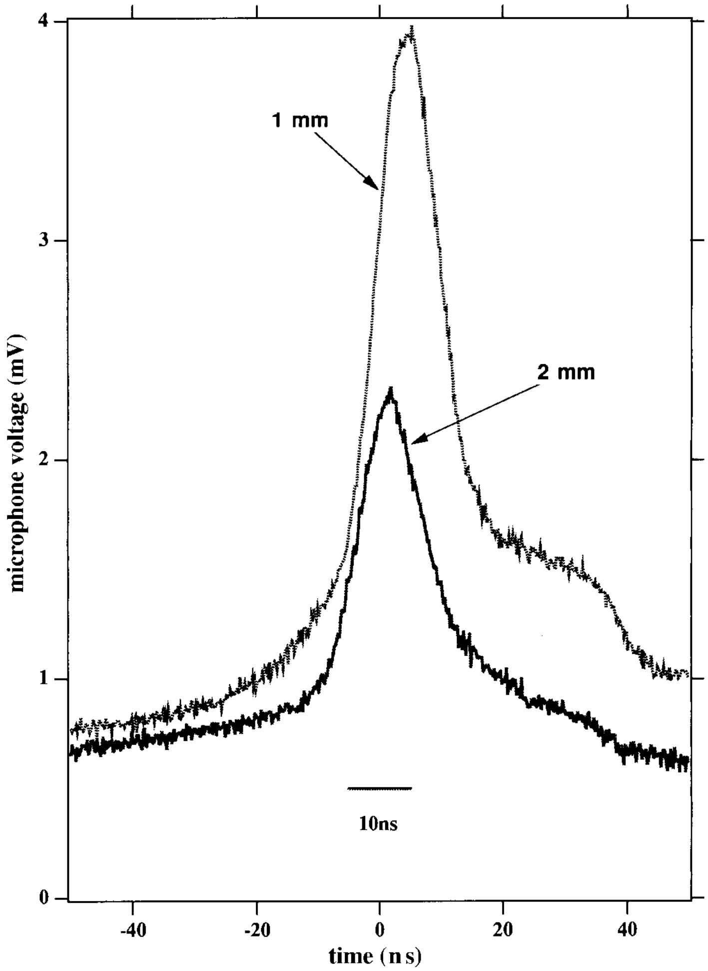

Figure 5. Sound pulse emitted by the bubble as it collapses. Measurements were made at 1 mm and 2 mm from the bubble and the fundamental drive frequency was filtered. The sensitivity of the microphone is 3.6 mV atm^{-1}. The measurements are limited by the bandwidth of the detector. The pulse clearly has acoustic frequencies above 30 MHz.

the bubble motion hands over to some light emitting mechanism at the point of maximum implosion.

Various models for the origin of the light have been proposed. The goal appears in each case to match the fact that the spectrum is broad band, displays no lines and increases into the ultraviolet (Hiller *et al.* 1992). Suggestions include Bremsstrahlung from thermal motion inside a submicron sized plasma (Wu & Roberts 1993), accelerated zero point motion (Casimir radiation) from an interface where the dielectric constant is rapidly changing (Schwinger 1993), and collisionally induced accelerations of molecular or atomic dipoles (Frommhold & Atchley 1994).

Although energy which is injected into the fluid at long wavelengths in the hydrodynamic regime focuses down to microscopic scales before coming out as light it remains to be seen whether Planck's constant plays a role in sonoluminescence.

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References

Barber, B. P. & Putterman, S. J. 1991 Observation of synchronous picosecond sonoluminescence. *Nature* **352**, 318–320.

Barber, B. P. & Putterman, S. J. 1992 Light scattering measurements of the repetitive supersonic collapse of a sonoluminescing bubble. *Phys. Rev. Lett.* **69**, 3839–3842.

Barber, B. P., Löfstedt, R. & Putterman, S. J. 1991 Sonoluminescence. *J. Acoust. Soc. Am.* Suppl. 1, S1885.

Barber, B. P., Hiller, R., Arisaka, K., Fetterman, H. & Putterman, S. J. 1992 Resolving the picosecond characteristics of synchronous sonoluminescence. *J. Acoust. Soc. Am.* **91**, 3061–3063.

Barber, B. P., Wu, C. C., Löfstedt, R., Roberts, P. H. & Putterman, S. J. 1994 Sensitivity of sonoluminescence to experimental parameters. *Phys. Rev. Lett.* **72**, 1380–1383.

Barber, B. P., Weninger, K., Löfstedt, R. & Putterman, S. J. 1995 Observation of a new phase of sonoluminescence at low partial pressure. *Phys. Rev. Lett.* **72**, 5276–5279.

Frenzel, H. & Schultes, H. 1934 Luminescenz im ultraschallbeschikter wasser. *Z. Phys. Chem.* B **27**, 421–424.

Frommhold, L. & Atchley, A. A. 1994 Is sonoluminescence due to collision-induced emission. *Phys. Rev. Lett.* **73**, 2883–2886.

Gaitan, D. F. 1990 Ph.D. thesis, University of Mississippi.

Gaitan, D. F., Crum, L. A., Church, C. C. & Roy, R. A. 1992 Sonoluminescence and bubble dynamics for a single cavitating bubble. *J. Acoust. Soc. Am.* **91**, 3166–3183.

Greenspan, G. & Nadim, A. 1993 On sonoluminescence of an oscillating gas bubble. *Phys. Fluids* A **5**, 1065–1067.

Hiller, R., Putterman, S.J. & Barber, B. P. 1992 Spectrum of synchronous picosecond sonoluminescence. *Phys. Rev. Lett.* **69**, 1182–1184.

Hiller, R., Weninger, K., Putterman, S. J. & Barber, B. P. 1994 Effect of noble gas doping in single-bubble sonoluminescence. *Science* **265**, 248–250.

Hiller, R. & Putterman, S. J. 1995 Observation of isotope effects in sonoluminescence. *Phys. Rev. Lett.* **75**, 3549–3551.

Löfstedt, R., Barber, B. P., Hiller, R. & Putterman, S. J. 1992 Limitations of the hydrodynamic theory of cavitation induced sonoluminescence. *Acoust. Soc. Am.* Suppl. 1, **91**, S2331.

Löfstedt, R. & Putterman, S. J. 1993 Toward a hydrodynamic theory of sonoluminescence. *Phys. Fluids* A **5**, 2911–2928.

Löfstedt, R., Weninger, K., Putterman, S. J. & Barber, B. P. 1995 Sonoluminescing bubbles and mass diffusion. *Phys. Rev.* E **51**, 4400–4410.

Putterman, S. J. 1995 Sonoluminescence: sound into light. *Sci. Am.* (February), 32–37.

Rayleigh 1917 On the pressure developed in a liquid on the collapse of a spherical cavity. *Phil. Mag.* **34**, 94–98.

Schwinger, J. 1993 Casimir light: the source. *Proc. Nat. Acad. Sci. USA* **90**, 2105.

Weninger, K., Hiller, R., Barber, B. P., Lacoste, D. & Putterman, S. J. 1995 Sonoluminescence from single bubbles in nonaqueous liquids: a new parameter space fo sonochemistry. *J. Phys. Chem.* **99**, 14 195–14 197.

Wu, C. C. & Roberts, P. H. 1993 Shock wave propagation in a sonoluminescing gas bubble. *Phys. Rev. Lett.* **70**, 3424.

Discussion

M. S. Longuet-Higgins (*Institute for Nonlinear Science, University of California, USA*). Dr L. A. Crum (Applied Physics Laboratory, University of Washington, Seattle, USA) has brought to my attention some interesting experimental results concerning single-bubble sonoluminescence, as described by Professor Putterman.

The experiment was performed in a high-flying aircraft (a pressurized DC 747) which executed a sinusoidal flight-path such that the effective value of gravity varied form 0 to $2g$ periodically. Apparently the luminosity from a single bubble suspended acoustically near an antinode was strongly correlated with the effective value g^* of gravity, being greatest when g^* was greatest. This variation in luminosity was reproduced on two separate flights.

The result suggests the possible importance of some assymetry in the initial conditions. Any asymmetry would be greatly magnified during the course of bubble collapse and could lead to inward microjets.

S. J. Putterman. The properties of sonoluminescence also depend upon the ambient pressure, which varies considerably during the transition from g to micro g.

J. T. Stuart (*Mathematics Department, Imperial College of Science, Technology and Medicine, London, UK*). Would Professor Putterman comment on the sensitivity to the presence or otherwise of argon, or to the doping with noble gases.

S. J. Putterman. We are unable to explain these remarkable results.