How can sound be transformed into a brief flash of light? Recent experiments have provided new insights into this remarkable phenomenon, but its cause is not yet fully understood

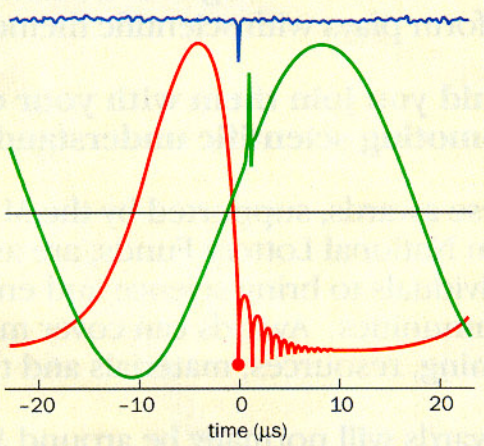
# Sonoluminescence: the star in a jar

### Seth Putterman

THE acoustics lab at the University of California in Los Angeles has seen a dramatic change in the past eight years. The transducers, microphones and amplifiers that are normally used for studying sound have now been joined by spectrometers, femtosecond lasers, the fastest oscilloscopes and photomultiplier tubes, as well as the most accurate time-interval meters. Equally important, but of a lesser technology, there is also equipment to purify water, and manifolds to prepare and control the gas content of fluids.

The phenomenon that demands this unusual confluence of equipment is sonoluminescence: the transformation of sound into light by the extraordinarily nonlinear pulsations of a gas bubble trapped in a fluid. Although sonoluminescence can be produced with equipment costing a couple of hundred dollars and can be detected by the eye for free, its measurement requires a set-up costing hundreds of thousands of dollars.

**1 Sound into light**



During a single cycle of the sound field, the pressure exerted on the bubble (green) follows a sinusoidal pattern. The bubble radius (red) expands during the rarefaction part of the sound field and collapses during the ensuing compression. At the minimum radius, a photomultiplier trained on the bubble records a flash of light (blue). The implosion also generates an outgoing pulse of sound detected by a microphone about 1 mm from the bubble, as shown by the spike on the sound wave. The time delay between the collapse and this spike is due to the finite speed at which sound propagates in water.

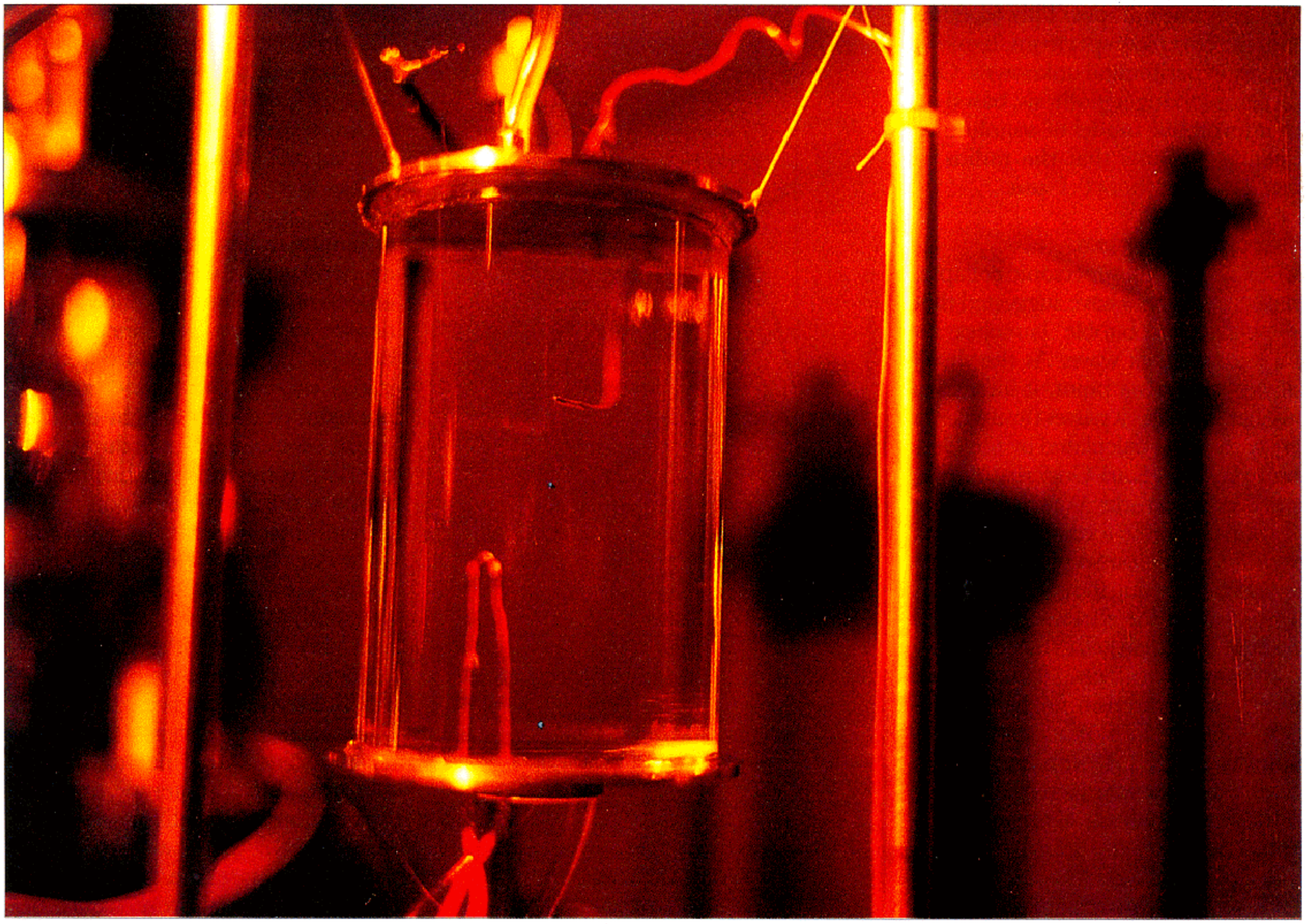
The two blue dots in the photo on the opposite page are due to sonoluminescence. In this typical arrangement, a cylindrical cell is formed from a glass tube with brass endcaps and filled with water. Piezoelectric ceramic transducers mounted on the endcaps vibrate when driven with an oscillating voltage and generate a sinusoidal sound field in the water. The frequency of the sound is 25 KHz, matching a radially symmetric "breathing" resonance of the water-filled cell. Frequencies above 20 KHz cannot be heard by the human ear and so are the most comfortable to work with.

Also shown in the photo is a small heater made from toaster wire. When a current passes through the heater, it boils the nearby water and forms vapourous cavities. Before disappearing, the cavities fill with whatever gas has been dissolved into the water. The Bernoulli pressure that a strong sound field exerts on a bubble gives rise to a force that prevents the bubble from floating away. Instead the force yanks the bubble to the centre of the resonator, where the acoustic pressure is at a maximum. However, like the construction of musical instruments the application of acoustic fields is to a large extent an art, and it is difficult to predict the spatial dependence of the vibrational modes in even this relatively simple resonator. The photo also shows that a second bubble has been trapped near the bottom endcap.

Once a bubble has been positioned by the sound field, the successive rarefactions and compressions cause it to pulsate rapidly (figure 1). During a rarefaction, when the pressure goes negative, the radius of the bubble expands from its equilibrium value of about 4 μm to a maximum value – about 40 μm in this case. The ensuing compression causes the bubble to collapse suddenly in a process that was first described by Rayleigh in 1917. The collapse reaches supersonic velocities and is stopped only when the gas inside the bubble is compressed to its van der Waals hard-core radius of 0.5 μm. At this moment, the flash of light due to sonoluminescence is emitted. The implosion also launches a pulse of sound when the bubble reaches its minimum radius.

## Experimental evidence

The resurgence of interest in sonoluminescence is largely due to the properties of the light flash, as measured by Bradley Barber and Robert Hiller at UCLA in the early 1990s. They found, for instance, that the spectrum of emitted light is broad band with no discrete lines, and that it extends from the infrared to the ultraviolet (figure 2). The photons emitted have energies of up to 6 eV, equivalent to temperatures of at least 70 000 K, indicating that the region generating the sonoluminescence must be very hot and highly stressed.



A snapshot of sonoluminescence – this photograph by Ed Kashi captures the light emitted from two isolated sonoluminescing bubbles in a water-filled cylinder, and shows exactly what your unaided eyes would observe in the laboratory. The photograph was taken by first exposing the film for long enough to record the deep blue light from the bubble – about 30 000 light flashes are generated per second, so an exposure of a few seconds is sufficient. These bubbles are filled with xenon to yield a bright bubble. The same frame of film is then exposed a second time to record the experimental apparatus. It is interesting that the dots of light are so precise. The bubbles remain at exactly the same location, despite all of the nonlinear pulsations that lead to the emission of light.

Photons with even higher energies might be generated, but these do not travel through water and cannot be detected. Indeed, one of the key unsolved puzzles in sonoluminescence is the spectrum beyond the cut-off imposed by water.

Another amazing property of the light is that the duration of the flash is measured in picoseconds, making sonoluminescence the only way to produce such short flashes without the use of a laser. The duration of each flash depends on the gas dissolved in the bubble (figure 3). If the water is degassed so that it contains only 3% of its usual content of dissolved air, the flash lasts for 40 ps. But if the water contains dissolved xenon, the flash can last for as long as 350 ps.

A typical light flash contains $10^5$–$10^7$ photons that are emitted uniformly in all directions. The intensity of the flash depends on the strength of the sound field, the temperature of the water, the external ("atmospheric") pressure, the gas–water mixture and other impurities that are sometimes difficult to control or diagnose.

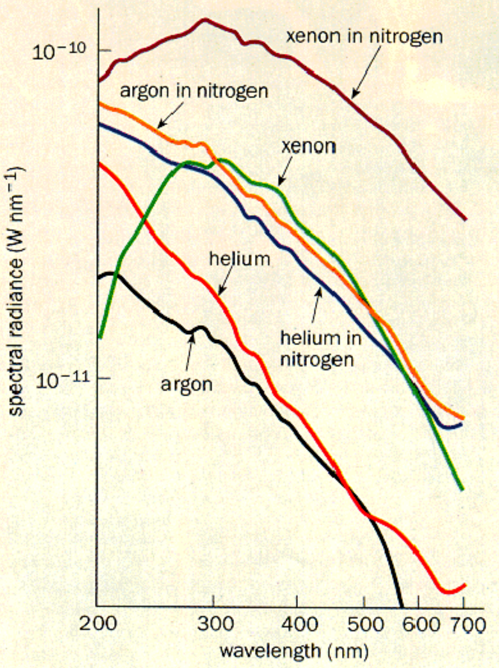
Just before the light is emitted, the bubble is collapsing at about 1.4 km s$^{-1}$, over four times the speed of sound in the gas (figure 4). We have measured the change in radius at times within 50 ns of the bubble's minimum size – less than one-tenth of the width of the dot at the bottom of the collapse shown in figure 1. These measurements were made by probing the bubble with precisely timed flashes of light from a femtosecond laser and recording the fraction of scattered light with a phototube. The fraction of light scattered from the bubble's surface depends on the square of the bubble radius.

Not only is the collapse supersonic, but the bubble "bounces" at the minimum radius with an acceleration that exceeds $10^{11}g$, where $g$ is the acceleration due to gravity. These same measurements show that the light flash and the minimum radius of the bubble coincide to within 0.5 ns.

After such a powerful implosion, one would reasonably expect that the bubble would shatter and disappear. To the contrary, the bubble is immediately ready for the next cycle of sound and the entire sequence, including the flash of light, is repeated. Moreover, these implosions can repeat with a clock-like synchronicity, such that the variation in time between flashes is less than 50 ps. This is surprising, since the system is also highly nonlinear. If we define a nonlinear coefficient, $G$, to be the ratio between the accelerations reached at the minimum and maximum radii, then $G = 10^6$ for sonoluminescence (and, of course, $G = 1$ for a harmonic oscillator). This nonlinear motion focuses the ambient acoustic energy by a factor of one trillion to produce the flash of light. It is remarkable that such an extremely nonlinear system can also be so regular, without a hint of chaos.

Although it is easy to see the dot from which sonoluminescence originates, no-one has yet measured the size of this light-emitting hot spot. This, together with the complete spectrum, is one of the key experimental unknowns. Our current working hypothesis, based on an interpretive model of sonoluminescence, suggests that the radius of the hot spot is about 0.1 µm.

**2 Spectrum of sonoluminescence**



The light emitted due to sonoluminescence was measured as a function of wavelength for various gas mixtures dissolved into water at 150 torr and at room temperature. In each case, the spectrum is smooth and contains no discrete lines. The spectrum at wavelengths below 200 nm has not been obtained because such high-energy photons cannot propagate through water.

## A shock to the system

Motivated by the supersonic collapse, many researchers have suggested that the collapsing bubble launches an imploding shock wave. As the shock focuses towards a point, it intensifies and heats the gas it passes through – in a way that was calculated by the German physicist Karl Guderley during the Second World War. When the shock reaches the focus, it "bounces" back and travels outwards through the heated gas, making the gas even hotter. According to the shock-wave model, the light is emitted at this moment.

To complete this model, there must also be a mechanism for light emission. Cheng-Chin Wu and Paul H Roberts of UCLA propose that the shock wave heats the gas enough for it to become ionized. The electrons released during the ionization then emit light as they collide with the ions.

This model can explain several experimental observations. First, it suggests that the bubble needs to collapse at about four times the speed of sound in the gas to launch the shock wave, which is consistent with experiments. It also requires that sonoluminescence and the minimum radius should be reached at the same time. But the most interesting prediction is the fact that the spectrum of light emitted by such free thermal electrons is also broad-band without discrete lines. Indeed, at large wavelengths, $\lambda$, the spectrum is proportional to $\lambda^{-2}$, and the sonoluminescence spectra are reasonably consistent with this law. To reach the temperatures needed to emit 6 eV photons, the model also predicts that the radius must become as small as 0.1 µm – our proposed but not measured size of the hot spot.

This model also appears to be qualitatively consistent with our recent observation – following initial evidence from Wolfgang Eisenmenger and colleagues at the University of Stuttgart – that the duration of the light flash is the same at all wavelengths, within an uncertainty of just a few picoseconds. If the material inside the bubble heated up and cooled down smoothly, the gas would spend more time at lower temperatures. The light emission would then last longer for low-energy (red) photons than for ultraviolet photons. Instead, these measurements suggest that sonoluminescence originates from a medium that has been transformed into a high-energy phase. The light switches on at the transition to this phase – which, according to the plasma model, happens when the gas is hot enough to ionize the gas and liberate the electrons – and all of the colours switch off at the same time as the material cools below the phase transition. Taken together, the numbers given above suggest that the plasma generating the sonoluminescence has a particle density of $10^{24}$ $cm^{-3}$ and that the net free charge of the hot spot is $10^9$ $e$, where $e$ is the charge on an electron.

A valuable feature of this plasma model is its prediction that the hot spot is small compared with the minimum radius. But most exciting is its consistency with the universal aspects of sonoluminescence. In particular, all gases have very similar broad-band spectra. Moreover, the initial experimental conditions do not seem to affect the light-emitting phase – for example, the duration of the flash is the same for helium and xenon at the same intensity.

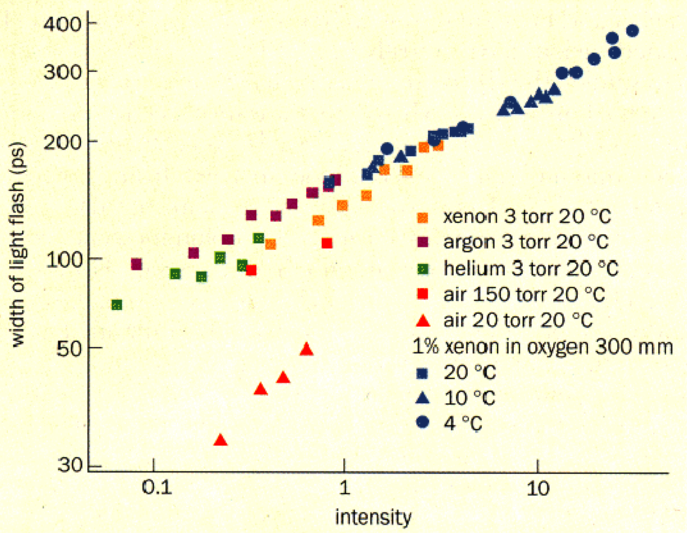
Tempting though this model may be, it does have a number of shortcomings. First, it does not determine the minimum radius of the shock front. In Guderley's theory the shock reduces to zero radius and reaches infinite temperature before bouncing back. In reality there is a minimum shock radius and a maximum temperature, but the temperature of sonoluminescence – like its spectrum – is unknown. This is a question with practical importance. If the bubble contained deuterium, it would become hot enough to generate fusion if the radius became as small as 10 nm.

The prospect of using a bubble to produce fusion is supported by a remarkable aspect of the rapidly changing radius (figure 4). The acceleration of the bubble points towards the centre, which means that the heavier water accelerates into the lighter gas. In this case the implosion is mechanically stable against shape instabilities, a situation that is obtained for free with sonoluminescence but which is difficult and costly to achieve in directly driven inertial-confinement fusion.

It is frustrating that experiments have not been able to measure the minimum shock radius, and that the simple shock-wave model does not answer this essential question. Since sonoluminescence is accompanied by the formation of a singularity, almost any physical process will be amplified and provide some limiting size to the shock focusing. Indeed, many researchers have calculated the effects of mass diffusion, thermal diffusion, viscosity, evaporation–condensation, surface tension, corrections to the equation of state, dissociation and chemical reactions on sonoluminescence. The fact that sonoluminescence is observed at all, despite the fact that so many large effects are competing at the same time, suggests that the underlying paradigm is not yet understood.

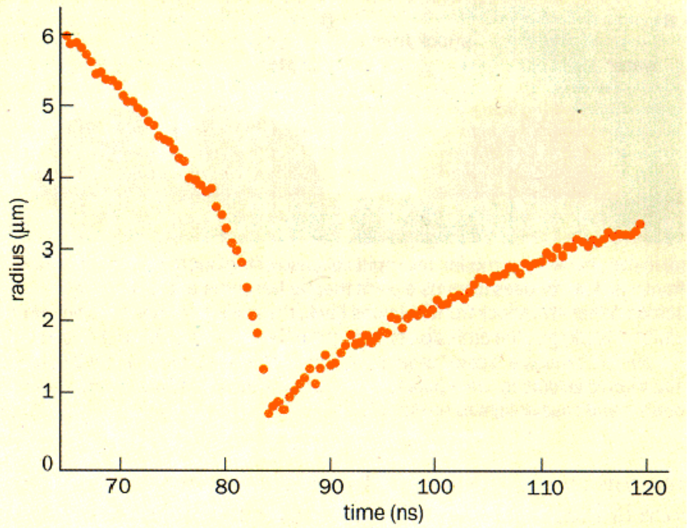
The shock-wave theory – or indeed any other theory for sonoluminescence – must explain the key role played by noble gases. Pure oxygen and nitrogen do not work, but the bubble will light up when just 1% of a noble gas is added. Barber has also noted that a bubble formed from air dissolved in water at 300 torr contains the same amount of argon as a bubble formed from pure argon in water at 3 torr. Furthermore, the

**3 Light flashes last for picoseconds**



The duration of the light flash depends, among other things, on the total intensity of light emission, the gas–water mixture and other experimental conditions. The shortest flashes are recorded when air is dissolved into water at low partial pressure. The longest flashes are achieved with the brightest bubbles, which contain mixtures of xenon and oxygen in cold water. The intensity axis is normalized to unity, which corresponds to the intensity of an air bubble in water at room temperature and a partial pressure of 150 torr. We obtained the measurements using time-correlated single-photon counting, a technique that was first adapted to sonoluminescence by Bruno Gompf, working with Wolfgang Eisenmenger at the University of Stuttgart in Germany. Our early experiments had to rely on measuring the response time of a fast photomultiplier tube and could only determine an upper bound.

**4 Minimum radius and maximum compression**



As the bubble approaches its minimum radius, it collapses at over four times the speed of sound in the gas. The smallest radius is reached when the van der Waals forces between the molecules do not allow them to get any closer. The measurements were made using pulsed Mie scattering, a technique developed by Bradley Barber and Keith Weninger.

light-emitting properties of these bubbles are similar, suggesting that a sonoluminescing bubble containing a mixture of gases only produces light from the atomic noble gas and expels the diatomic molecules from the hot spot. This idea does not withstand further scrutiny, however, indicating that the situation is more complicated than this scenario. For example, mixtures of hydrogen with noble gases do not display this doping effect. Indeed, none of the theoretical models proposed so far can account for all of the sensitive and intricate effects of gas composition on sonoluminescence. Yet another shortcoming of the shock-wave model is that it cannot explain the bubble radius and the range of acoustic pressures at which sonoluminescence occurs.

### Alternative theories and tests

Many other theories have been suggested to explain sonoluminescence. For example, Michael Brenner of the Massachusetts Institute of Technology in the US, and Detlef Lohse and colleagues at the University of Marburg in Germany have attempted to explain the sonoluminescence phase diagram and the effect of noble gases in terms of chemical reactions, mass diffusion and shape instabilities. Paul Roberts and other colleagues in my lab dispute this particular theory but, rather than dwelling on the reasons why, I would like to propose an overarching standard for all theoretical papers: they should be judged on whether the theory makes a prediction that is almost as interesting as the discoveries they try to explain.

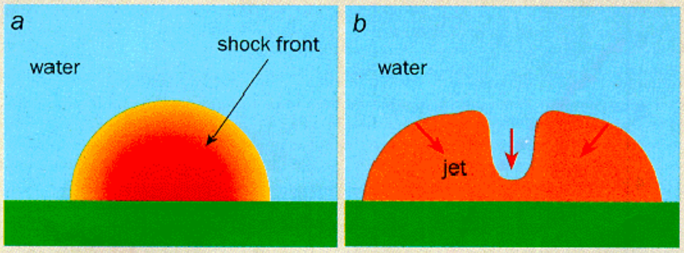
One fundamental aspect of physics that has unfortunately not been shown to play a role at the singularity is quantum mechanics. It would be most remarkable if Planck's constant were to control some macroscopic aspect of low-frequency sound travelling through a fluid at room temperature. For this reason, a wave of excitement was generated when Claudia Eberlein, now at Sussex University in the UK, and the late Julian Schwinger suggested independently that radiation can be produced in accelerating dielectrics as a result of changes in the so-called quantum zero-point motion. However, many researchers have since pointed out that the singularity associated with sonoluminescence is too small by many orders of magnitude for this aspect of quantum mechanics to kick in.

Perhaps the strongest alternative to the shock-wave theory is the "jet" model, developed independently by Andrea Prosperetti of Johns Hopkins University in Baltimore and Michael Longuet-Higgins of the University of California in San Diego. In this picture the collapsing bubble becomes unstable against the formation of a spike of water that is propelled through the centre of the bubble at supersonic velocities. This interesting idea has firm roots in the well established science of underwater munitions. For example, depth charges do not destroy a submarine through the explosion itself, but by the jet of water formed in the inward collapse of the cavity it creates. The jet, which is attracted to the nearest solid boundary, punches a hole in the hull of the submarine.

Indirect evidence for the jet model comes from our observations of sonoluminescence from hemispherical bubbles attached to solid boundaries. These bubbles also display picosecond flashes and have spectra very similar to those of isolated spherical bubbles. Jets are expected to exist in collapsing wall bubbles because they are blamed for cavitation damage to propellers and turbines. Since the properties of wall bubbles and spherical bubbles are so similar, it seems logical to suggest that jets also lead to sonoluminescence in spherical bubbles. On the other hand, it could turn out that cavitation damage due to wall bubbles could in some cases be caused by imploding hemispherical shocks, rather than jets (figure 5).

To distinguish between the two models and find out more about sonoluminescence, a key parameter to measure will be the size of the light-emitting hot spot. Steve Trentalange of UCLA and Sanjeev Pandey of Ohio State University suggest

**5 Comparison of the shock and jet models**



(*a*) In the shock-wave model, the rapid collapse of the bubble creates a shock front that first focuses towards a point that we believe is about 0.1 µm across. When the shock front bounces back, the gas is hot enough to become ionized, leading to the emission of light. (*b*) In the jet model, the implosion of the bubble causes a "spike" of water to form that propels into the bubble at supersonic velocities. The spike slams into the water on the other side of the bubble and causes light to be emitted.

that this could be achieved by measuring how the intensity correlations of the light flash varies with angle, in the same way that Hanbury-Brown and Twiss determined the radius of a star. Indeed, the solid angle subtended by the bubble at a photodetector is about the same as that of a nearby star.

Another possible method for obtaining the size of the hot spot is to measure the Thomson scattering from free electrons inside the bubble. Such a measurement would also provide a diagnostic for the presence of a plasma, since it has not yet been established that a plasma exists inside the bubble at the moment of light emission.

It is also vital to measure the peak temperature reached in sonoluminescence. While Thomson scattering might provide some clue, a search for fusion in a deuterium bubble would provide an upper bound of about 1 KeV – or else an amazing discovery. Unfortunately nature refuses to make this an easy experiment, since deuterium bubbles last for at most a minute before fading away.

## From then until now

The history of research on sonoluminescence has gone through many twists and turns. Its first mention was perhaps a miracle reported in the original version of Exodus (20:18), "And all the people saw the sounds", but this was removed in later translations. Frenzel and Schultes of the University of Cologne actually discovered sonoluminescence in 1934, when they made meticulous measurements of the light emitted from a cloud of cavitating bubbles. However, in the paper reporting their discovery Frenzel and Schultes also mention their plans to turn their attention to more important issues.

Over 30 years later Frank Petersen and Thomas Anderson at Northwestern University in Illinois observed luminescence from cavitation bubbles generated in water flowing through a Venturi tube. Amazingly, their observations led them to claim that the individual flashes of light were shorter than a nanosecond. On tracking this work forward, we find only one research citation – to a highly critical comment from the cavitation establishment. It said that picosecond timescales are an obvious consequence of the theories of bubble motion, and so the experiments were not interesting. This was 1966, and Petersen and Anderson had discovered the fastest man-made source of light. One can only wonder why they did not mention this in their paper or in their timid reply, and another exciting line of research on luminescence from cavitation bubbles quickly died out.

I became interested in sonoluminescence in 1988, when I heard about the phenomenon from Tom Erber. Bradley Barber and I initiated a research programme at UCLA and I visited various labs – mostly funded by the US Navy – that had studied the allied problem of cavitation for many years.

At the University of Mississippi I was surprised to hear that Felipe Gaitan, working with Lawrence Crum, had discovered that sonoluminescence could be realized from a single isolated bubble. I enthusiastically told them that they had stumbled upon the hydrogen atom of sonoluminescence, and encouraged them to measure the light emission. However, their efforts had not met with much excitement from the cavitation establishment, and they had already dismantled their apparatus. Gaitan and Crum realized that I was excited about sonoluminescence as a result of my views on the energy focusing and so, like gentlemen, they helped my colleagues and me to tune our existing set-up to the region where they had observed sonoluminescence from a single bubble.

When I look at what Bob Hiller calls the "star in a jar" I cannot help but marvel at the wonders and teases of nature. An externally imposed sound field with a low velocity and a long wavelength can spontaneously focus its energy to create a brilliant hot spot that is easy to see with the naked eye. Yet after years of staring at this bright bubble, we have not yet succeeded in measuring its temperature and we are only now closing in on its size.

## Further reading

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**Methods to produce single-bubble sonoluminescence**

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B P Barber, K Weninger, R Löfstedt and S Putterman 1995 Observation of a new phase of sonoluminescence at low partial pressure *Phys. Rev. Lett.* **74** 5276

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