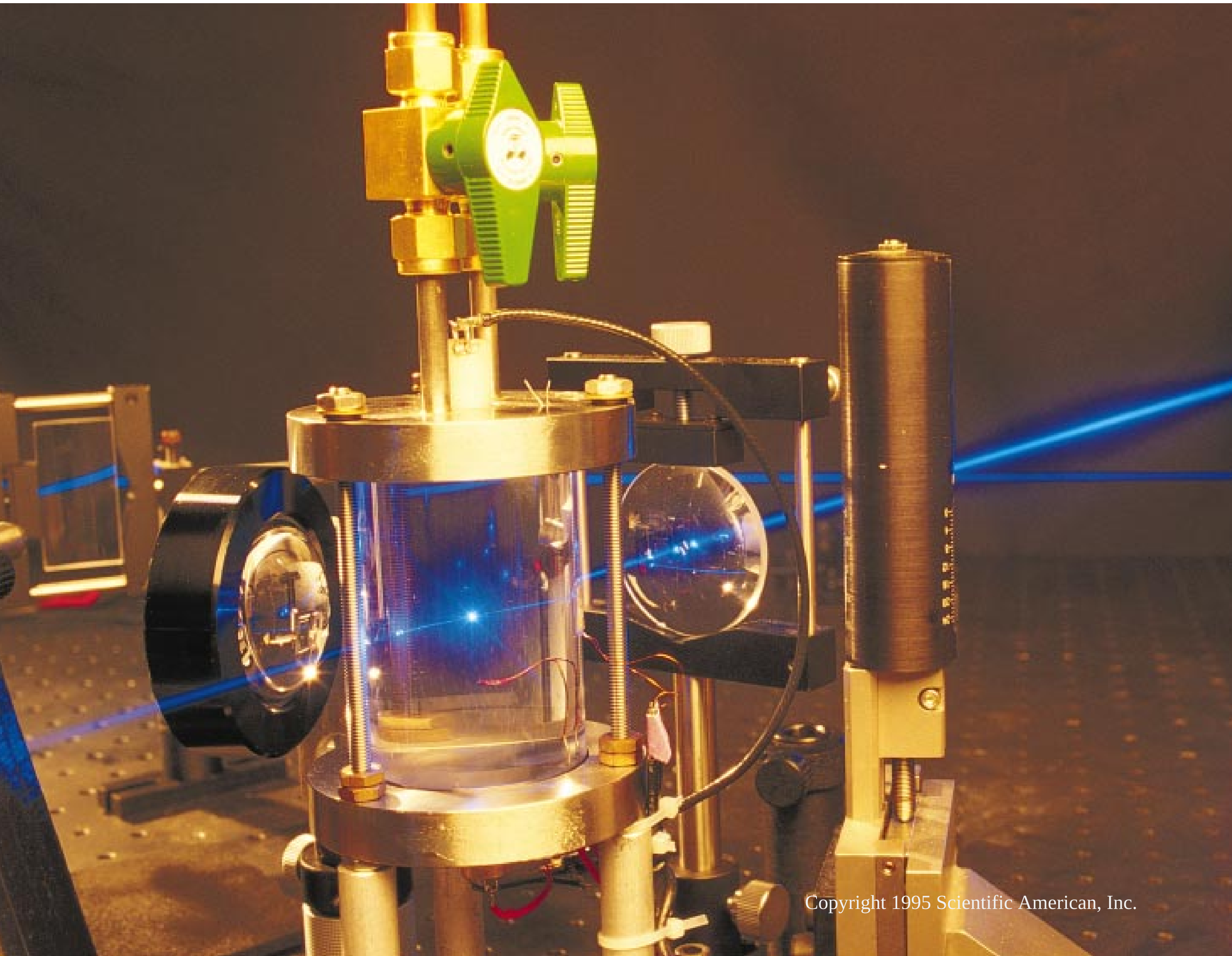
Sonoluminescence: Sound into Light

A bubble of air can focus acoustic energy a trillionfold to produce picosecond flashes of light. The mechanism eludes complete explanation

by Seth J. Putterman

Imagine you are riding a roller coaster. First, you chug up a long incline slowly. When you get to the top, your car free-falls, speeding up until it reaches the bottom of the drop, where the deceleration crams you into your seat. That sensation is what you would feel if you were riding a pulsating bubble of air trapped in water—except that the drop would reach supersonic speeds and at the bottom you would be crushed into your seat with a force equal to 1,000 billion times your weight.

Obviously, more than your stomach would react to such a ride. As for the bubble, it responds to the extraordinary force by creating a flash of light only a tiny fraction of a second long. The light



is mostly ultraviolet, which indicates that when the bubble's free fall stops, its interior becomes much hotter than the surface of the sun. A sound wave can make the bubble repeat this wild ride more than 30,000 times a second, so that the flashes burst out with clocklike regularity.

In sonoluminescence—as the process of converting sound into light is called—the bubble is concentrating the energy of the acoustic vibrations by a factor of one trillion. That is, the sound wave that drives the bubble is centimeters long, but the light is emitted from a region of atomic dimensions.

A detailed accounting of this inexpensive yet unusual illumination source remains elusive. The flashes are so brief that to measure the properties of light, we must use photodetectors that respond more quickly than those employed by high-energy physicists. (In fact, sonoluminescence is the only means of generating picosecond flashes of light that does not require expensive lasers.) The physical process by which sonoluminescence achieves such a huge focusing of energy may serve as a useful model for researchers seeking to develop controlled nuclear fusion. Current attempts to fathom the mysteries of sonoluminescence in my laboratory at the University of California at Los Angeles and in other institutions are generating new paradoxes faster than the existing questions can be answered.

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Skeptical Inquirer

I was actually quite incredulous of sonoluminescence when I first heard about it in the mid-1980s from my scholarly colleague Thomas Erber of the Illinois Institute of Technology. One day at the U.C.L.A. coffeehouse, he taunted me about my long-standing interest in fluid mechanics, focusing on the Navier-Stokes equations, which describe the flow of fluids. He asked, "If you think the Navier-Stokes equations are so great, then please explain to me how sound can be made into light." Based on my intuition, I replied that I did not believe sonoluminescence was possible. But he insisted that this effect had been documented some time ago. So along with Ritva Löfstedt, who was then a U.C.L.A. undergraduate, I went back through the old papers to see if sonoluminescence was for real.

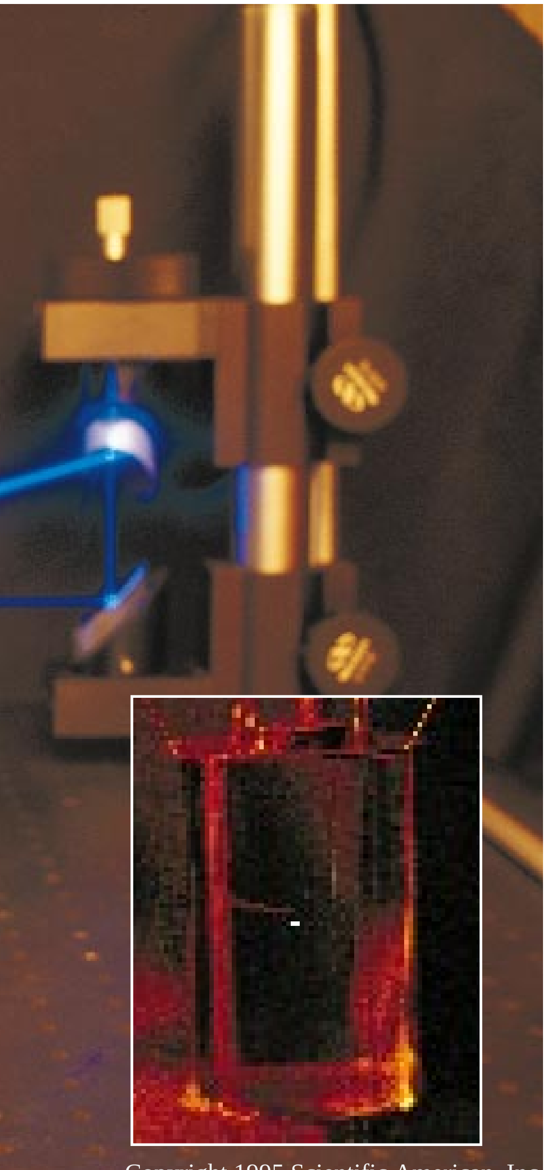
In the 1920s and 1930s, we learned, chemists working with loudspeakers developed for sonar systems during World War I came across an interesting phenomenon: a strong sound field could catalyze reactions that take place in an aqueous solution. A German scientist, Reinhard Mecke of the University of Heidelberg, then commented to his coworkers that the amount of energy needed for a chemical reaction is the same as that needed to excite the emission of light from an atom. So he suggested a search for such a signal. Soon afterward, in 1934, H. Frenzel and H. Schultes of the University of Cologne discovered sonoluminescence in a bath of water excited by acoustic waves.

Perhaps it was the common observation that one can generate a spark of light by touching a doorknob after walking on a carpet. Whatever their inspiration, Frenzel and Schultes explained the light emission in terms of *Reibungselektrizität,* or "frictional electricity." In their experiment the sound wave initiated the process of cavitation—the growth and collapse of bubbles in water. They pictured the bubbles' motion through the liquid as analogous to that of shoes shuffling on a carpet. The abrasion causes electrical charges to separate in the originally neutral media. A spark releases the built-up charge. Then they concluded their paper by saying they had more important matters to attend to.

Other researchers, seeking clues to the mechanism, proceeded to carry out spectral measurements of this new light source. These studies were inconclusive because of the transient nature of the phenomenon. The strong sound fields they used created clouds of bubbles that grew, collapsed and gave off light in an unpredictable and unsynchronized manner.

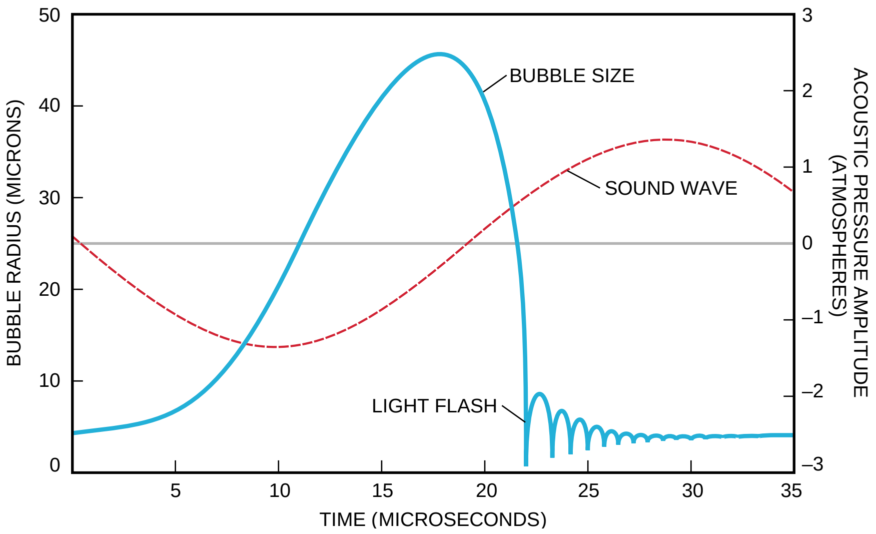
At U.C.L.A., Bradley P. Barber, a graduate student, and I became enthusiastic about characterizing and understanding the mechanism responsible for sonoluminescence. I learned that other investigators had just succeeded in trapping a single, light-emitting bubble in water that was partially degassed. They were D. Felipe Gaitan, now at the Naval Postgraduate School, and Lawrence A. Crum, now at the University of Washington. It seems that my enthusiasm for their advance far exceeded theirs. They had dismantled the experiment and abandoned this avenue of research. But they did show us how to adjust our apparatus to find single-bubble sonoluminescence.

So with a boiling flask from the chemistry laboratory, an oscilloscope from the undergraduate lab, my home stereo and a photomultiplier tube (light sensor) purchased with my credit card, we were off and running [see "The Ama-



ED KASHI; SETH J. PUTTERMAN AND ROBERT A. HILLER (*INSET*)

MAKING LIGHT OF SOUND is accomplished by a bubble of air trapped in a cylindrical flask of degassed water. Sound from speakers above and below the flask trap the bubble. A flash of light 50 picoseconds long emerges during the compression part of the acoustic wave. A laser measures the bubble size as it pulses in time with the sound. The light emission itself is rather faint (*inset*).



JARED SCHNEIDMAN

ROLLER-COASTER RIDE of a pulsating bubble lags slightly behind the expansion and compression of sound waves. The bubble expands to its maximum radius just after the acoustic pressure becomes negative. During compression, the bubble rapidly shrinks to less than one micron in radius and emits a flash of light. The bubble continues to swell and contract briefly before settling down.

teur Scientist," page 96]. For some of our initial work, we injected an air bubble into water with a syringe. Over the years we have refined our setup. Our current apparatus consists of a piezoelectric transducer on the top of a cylindrical flask filled with water. The transducer is a ceramic material that turns an oscillating voltage into a mechanical vibration and thereby sets up sound waves—alternating fields of compression and expansion—in the water. Submerged in the water is a small piece of toaster wire. When current flows through it, the wire heats up, boiling the water nearby. As a result, a bubble filled with water vapor forms. Before the vapor recondenses, air dissolved in the water flows into the pocket to create an air bubble.

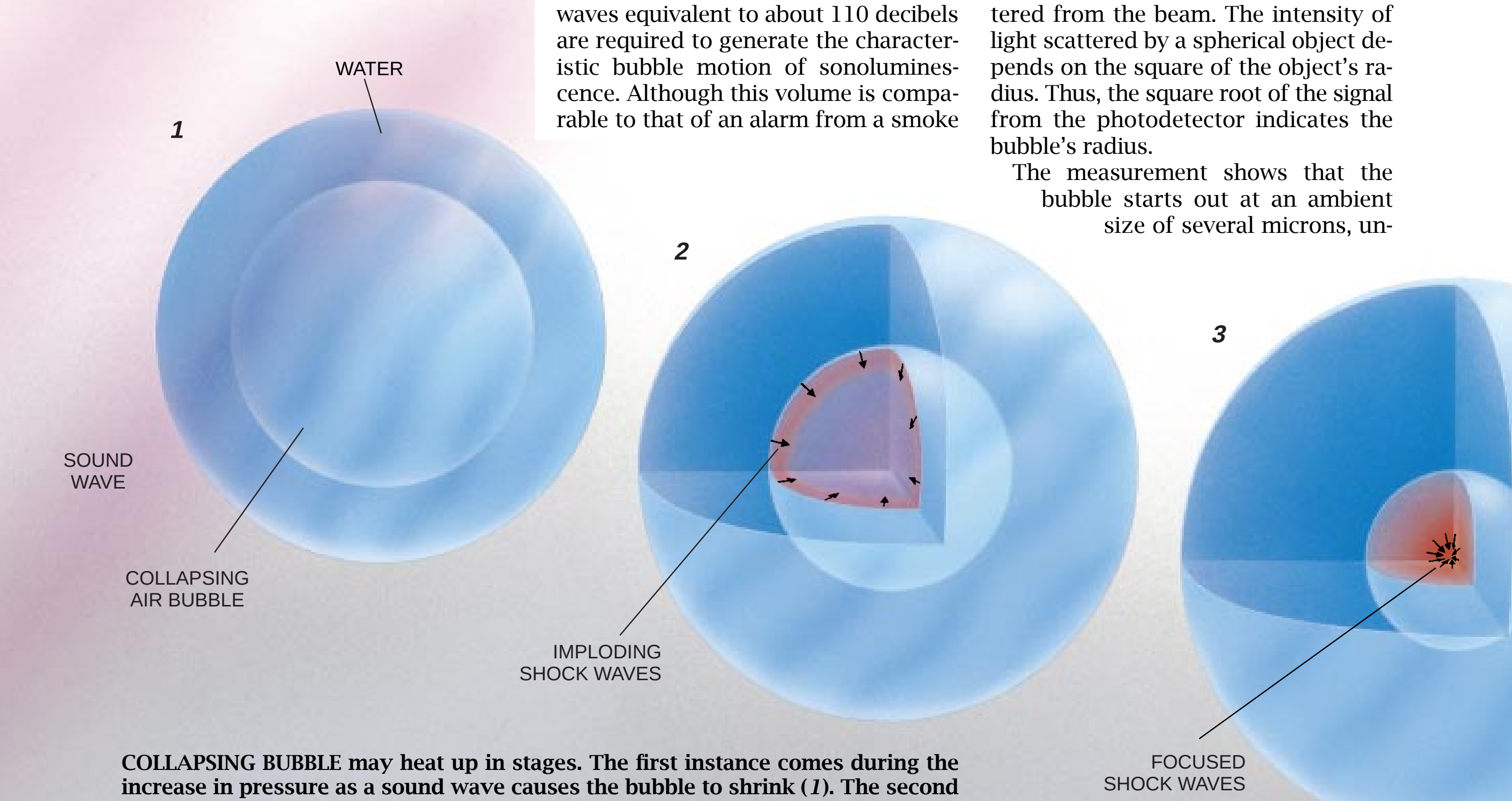
This bubble is then trapped at the center of the cylindrical flask, where the buoyancy force that would make the bubble rise to the top is balanced by the force of the sound waves. Acoustic waves equivalent to about 110 decibels are required to generate the characteristic bubble motion of sonoluminescence. Although this volume is comparable to that of an alarm from a smoke detector a few centimeters away, the frequency of the sound lies just beyond the range of human hearing.

Probing the Bouncing Bubble

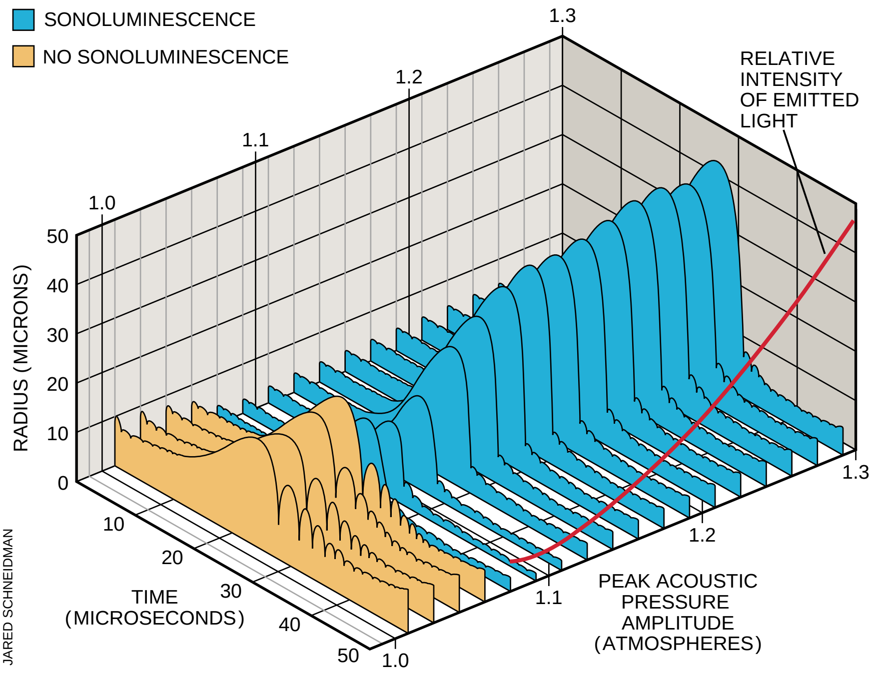
As physicists attempting to characterize sonoluminescence, our first goal was to identify the time scales involved in the process—specifically, the duration of the flash. We were amazed to find that such a measurement would require the use of the fastest known light sensors. Our analysis yielded an upper bound of about 50 picoseconds. We also found that the flashes came out with an incredible regularity. The timing between consecutive flashes, typically about 35 microseconds, varies by no more than 40 picoseconds.

To determine the radius of the sonoluminescing bubble, Barber shone a laser on it and measured the light scattered from the beam. The intensity of light scattered by a spherical object depends on the square of the object's radius. Thus, the square root of the signal from the photodetector indicates the bubble's radius.

The measurement shows that the bubble starts out at an ambient size of several microns, un-



COLLAPSING BUBBLE may heat up in stages. The first instance comes during the increase in pressure as a sound wave causes the bubble to shrink (*1*). The second occurs as the walls collapse at supersonic speeds, which in turn launches shock waves toward the bubble center (*2*). The shock further heats the interior. Even more heat is generated as the shock waves focus (*3*) and then explode out (*4*).



JARED SCHNEIDMAN

TRANSITION TO SONOLUMINESCENCE happens when the sound level reaches a critical state. The average radius of a bubble generally increases with a rise in acoustic amplitude. At the level at which sonoluminescence begins, however, the radius suddenly shrinks. The mechanism behind this transition is not understood.

til the expansion part of the sound field acts on it. Then the pressure drops, putting the fluid under tension and causing the bubble to swell to about 50 microns. The expansion continues until the sound field switches from rarefaction to compression.

At this point of maximum expansion, a near-vacuum has formed inside the bubble. That is because the volume of the bubble has greatly increased, but the number of molecules inside it has not changed. Atmospheric pressure, however, still acts on the outside of the bubble. The pressure difference between the inside and the outside leads to a catastrophic collapse. The bubble decreases from its 50-micron maximum radius to a size of about 0.5 micron. At that point, the surface of the bubble stops its inward rush as though it had suddenly slammed into a wall. It cannot become any smaller because of the repulsive force between the gas atoms and molecules. (We say at this point the size of the bubble is determined by the van der Waals forces of the hard core of its contents.) The light flash comes out as the bubble decelerates through its minimum radius [*see top illustration on opposite page*]. After the light emission, the bubble elastically bounces in size a few times and then sits dead in the water waiting for the next helping of sound.

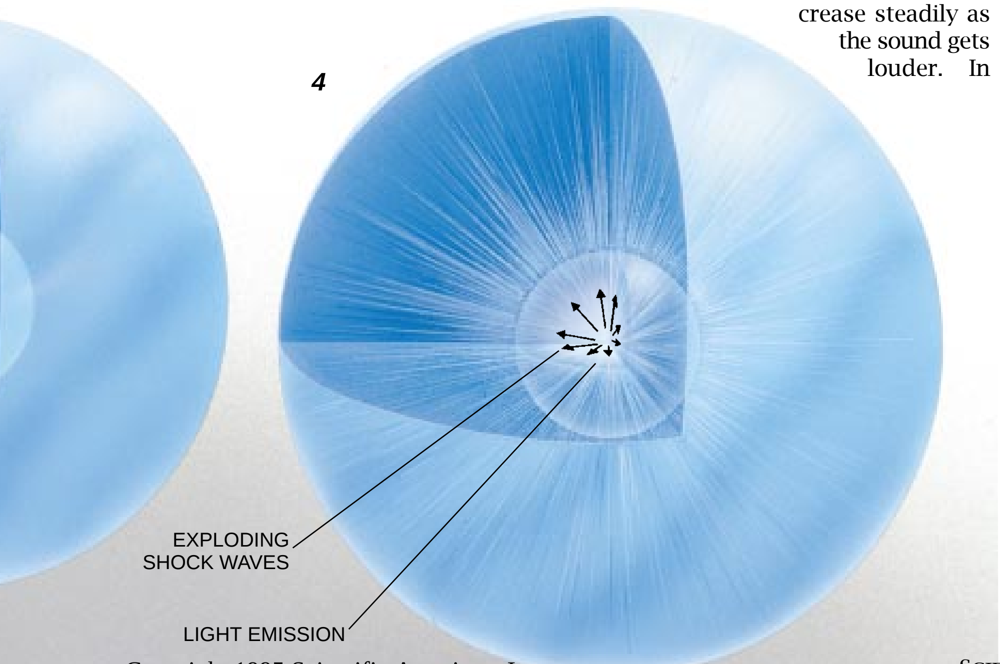
Although experiments can measure the size of the bubble, no theory can explain how those particular radii come about. The size of a bubble depends on the amount of gas trapped inside. Löfstedt, now one of my graduate students, and I are studying the mechanism whereby the gas dissolved in the surrounding water diffuses into the bubble. When the bubble is large, the pressure inside it is low; therefore, gas flows into it from the surrounding fluid. When the bubble is small, the reverse occurs. The balance between inflow and outflow of air molecules determines the average bubble size.

The radius of a bubble driven by a weak sound field seems to follow the predictions of this model. But applying the same reasoning to a high-amplitude, light-emitting bubble leads to a contradiction with the data [*see illustration above*]. The average radius of the bubble should be seen to increase steadily as the sound gets louder. In practice, however, this relation has an unusual discontinuity just as sonoluminescence sets in: the average radius suddenly decreases for a moment. Beyond that point, it rises with sound amplitude again. Some new (and as yet unknown) mass-flow mechanism must determine the sonoluminescent state.

Torrid Interior

To the unaided eye, the faint blue glow of a sonoluminescing bubble looks like a star in the night sky. In 1991 my graduate student Robert A. Hiller determined how much of this radiated light lies in the visible part of the spectrum. He found that there is literally more to the spectrum than meets the eye. The results showed that the bubble emits more purple light than red and more ultraviolet than purple. We could not follow the spectrum beyond photon energies of six electron volts, corresponding to an ultraviolet light wavelength of 0.2 micron, because above those energies light cannot propagate through water. (For the same reason, we had to construct our flask from quartz rather than plain glass, which blocks ultraviolet light.) An energy of six electron volts corresponds to a temperature of 72,000 kelvins, so the interior of the bubble must be scorching indeed.

That a collapsing bubble of gas becomes very hot can be explained in terms of an everyday experience for res-



TOMO NARASHIMA

idents of southern California and the Alps. These people suffer through particularly torrid weather when the wind blows from higher elevations to lower ones. In southern California, a “Santa Ana” condition occurs when air from the high desert heats up by 15 degrees Celsius as it blows into the Los Angeles basin. The sudden temperature increase results from the work performed by the atmosphere on the desert air mass as the air drops in altitude by 5,000 feet on its way to the ocean. At lower altitudes, the barometric pressure is higher. If the pressure difference compresses the air before it has time to exchange its heat with the ocean or other cooler bodies, the air becomes adiabatically heated—that is, its temperature rises without the addition of any heat energy.

The hot spot realized in a collapsing bubble is astronomical even when compared to a sizzling day in California. The volume of a sonoluminescent bubble drops by a factor of one million as its radius decreases 100-fold. In the 1950s B. E. Noltingk and E. A. Neppiras of Mullard Electronic Research Laboratory in Surrey, England, calculated that the resulting adiabatic compression of the bubble interior leads to a temperature of up to 10,000 kelvins and pressures greater than 10,000 atmospheres. (The bubble surface does not vaporize, perhaps because the high rate of pressurization and heating takes place well inside the bubble.)

Had the revered English physicist Lord Rayleigh lived in southern California, his experience with the weather might have led him to predict sonoluminescence as part of the bubble research that he carried out in 1917. The Royal Navy hired him to help understand the causes of the degeneration of ship propellers. Rayleigh determined that the small bubbles of air created as the propeller sliced through the water were the culprits. The bubbles would corrode the propeller as they collapsed onto it with a force greater than 10,000 atmospheres. But in describing the motion of the bubbles, he assumed the collapse of a bubble obeyed Boyle’s law: in other words, he thought the temperature inside it remained constant. Had he realized that the collapse is so rapid that it is adiabatic, he surely would have predicted high temperatures and the associated light emission.

Exactly how would the high temperature produce light? According to researchers who study sonoluminescence and sonochemistry, the energy from the collapse is powerful enough to break apart molecules within bubbles. The dissociated molecules emit light as they recombine. This effect, referred to as chemiluminescence, was first presented by Virginia F. Griffing of Catholic University in 1952. It accompanies transient cavitation and has been used to initiate unusual chemical processes. An example is the fabrication of amorphous iron by Kenneth S. Suslick of the University of Illinois [see “The Chemical Effects of Ultrasound,” by Kenneth S. Suslick; SCIENTIFIC AMERICAN, February 1989].

Although adiabatic heating of a collapsing bubble provides an impressive mechanism for energy concentration, it

Shock Waves in Bubbles

In the past, researchers who studied sonoluminescence and sonochemistry associated the transient clouds of cavitating bubbles with hot spots that formed within each bubble. In this traditional model the energy focused by the collapse of the bubbles creates dissociated molecules that emit light as they recombine.

Yet the prevailing wisdom about transient cavitation cannot explain the strongly ultraviolet spectrum emitted by a single bubble synchronized to the sound field. Our measurements indicate that the bubble’s interior attains a temperature substantially higher than 10,000 kelvins. This value can be reached if the collapse of a single synchronized bubble is so fast and symmetrical that it launches a spherical shock wave into its interior. As the imploding shock wave of radius R_s focuses, its amplitude and speed increase. For this case, the solution to the equations of hydrodynamics takes the form

$$R_s = At^b$$

where A is a constant, time t is measured from the moment of focusing when $R_s = 0$, and b is 0.7 for air.

A Mach number is associated with every shock wave. This number represents the ratio of the shock velocity to the ambient speed of sound. The temperature behind a shock front is higher than that in front of it; the ratio of those temperatures is proportional to the square of the Mach number.

For an imploding air bubble, the Mach number approaches infinity as the shock front moves closer to the focal point, which means that a tremendous amount of heating takes place. Furthermore, when the shock hits the center and explodes outward, the molecules that were behind the shock are suddenly in front of it again. The hot molecules are hit a second time, and their temperatures go up by another factor of the square of the Mach number.

Temperatures that can be reached by this mathematical model are therefore unimaginably high. In reality, they are limited by the stability of the shock front. In the shock-wave model, sonoluminescence hinges on the shock front remaining spherical down to a radius of about 0.1 micron.

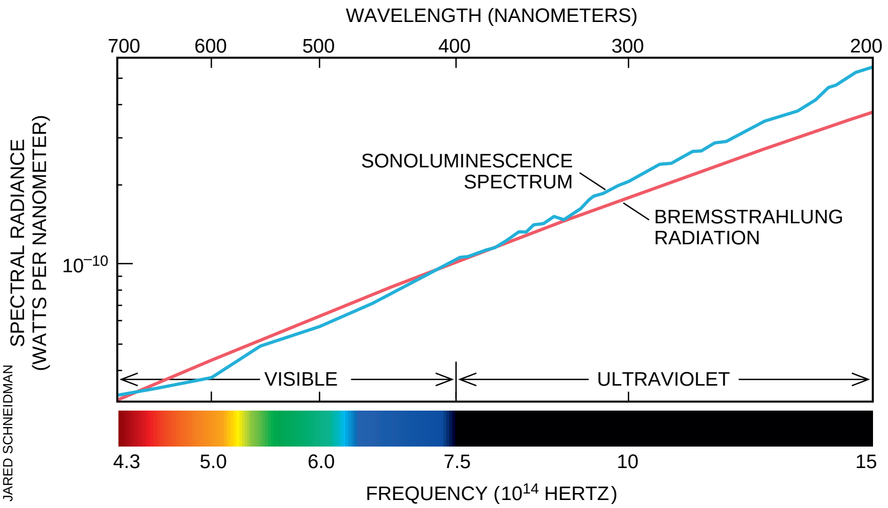
Spherical shock fronts played an important role in the design of nuclear weapons. British physicist Geoffrey I. Taylor used photographs and the corresponding expression for exploding shocks to obtain an unauthorized calibration of early hydrogen bomb tests. In the future, our understanding of how a sonoluminescing sound field generates such a beautifully spherical collapse could assist researchers at such institutions as Lawrence Livermore National Laboratory and the University of Rochester in designing improved versions of inertial confinement fusion. In this fusion process, huge lasers induce the implosion of a small pellet containing a mixture of the hydrogen isotopes deuterium and tritium. The spherical implosion is the key to reaching temperatures and densities sufficient to realize the fusion of these hydrogen nuclei to yield helium and neutrons.

There is a highly speculative chance that the comparison between inertial confinement fusion and sonoluminescence may indeed reveal a deeper similarity. If the sonoluminescent shock remains stable down to the incredibly small radius of 10 nanometers, then this tiny region would also reach temperatures appropriate to fusion. It is not hard to imagine many effects that would stand in the way of such an outcome—instability of the shock wave, thermal diffusion and radiation damping, to name a few. Given the shortcomings of current models, we bet that this issue cannot be decided by computer simulation. Only future experiments can tell if the interior of the bubble gets as hot as the interior of the sun.

cannot be the only or complete answer. Such heating alone would not be able to generate the largely ultraviolet spectrum we observed. Therefore, an additional stage of energy amplification must take place. Barber and I deduced a plausible mechanism. We realized that the supersonic speeds of the collapsing bubble could launch shock waves into the bubble's interior. Although the bubble's motion is arrested by the forces of the gas molecules against one another, the imploding shock wave could continue inward and further concentrate the energy of the collapse.

Our U.C.L.A. colleagues Paul H. Roberts and Cheng-Chin Wu also realized the potential importance of shock waves in sonoluminescence. They calculated the extent of the concentration. Building on a solution first developed in the 1940s by the German mathematician Karl G. Guderley of the Institute for Gas Dynamics in Braunschweig, they showed that the bubble's collapse could launch a shock wave into the bubble that becomes stronger as the shock implodes. The high temperature and pressure associated with this shock front become even more amplified when the converged shock subsequently explodes outward [*see box on opposite page*].

Typically, shock fronts are susceptible to instabilities that corrugate their surfaces, which thereby limit the extent of the implosion. If the inward-moving shock front launched by the bubble remains intact to a radius of 0.1 micron from the center of the bubble, the temperature near it would be 100,000 kelvins. This heat is about that required for the strongly ultraviolet spectrum we observed. If the shock front survives down to 20 nanometers, the temperature would reach one million kelvins, hot enough to make soft (relatively long wavelength) x-rays. Such photons do not propagate through water, so we do not know whether they are there. The possibility of getting weak x-rays from sound might seem far-fetched, and I am skeptical of such an outcome. Then again, I was quite doubtful of sonoluminescence in the first place.



JARED SCHNEIDMAN

SPECTRUM of sonoluminescence shows that most of the emitted light is ultraviolet. As pointed out by Paul H. Roberts and Cheng-Chin Wu of the University of California at Los Angeles, the signal compares closely with bremsstrahlung radiation—that is, light emitted by a plasma at 100,000 kelvins.

Noble Addition

Although the mechanism of sonoluminescence from a single bubble is difficult to explain, the phenomenon is easy to produce and modify. Despite being a robust phenomenon, it is highly sensitive to controllable experimental parameters, such as the intensity of the sound and the temperature of the water. For instance, the amount of light emitted with each flash increases by a factor of 200 as the temperature drops from 35 to 0 degrees Celsius. At 0 degrees, the bubble gives off about 10 million photons per flash.

The sensitivity to temperature suggested that we could learn more about sonoluminescence by changing other quantities. We attempted to find single-bubble sonoluminescence in liquids other than water, but without success. Because we could not change the driving fluid, we tried changing the gas in the bubble. This alteration entailed degassing the water by exposing it to a vacuum, a process that removes the dissolved air. Then we dissolved other gases of our choice into the water. Obviously, this procedure had to be carried out in an airtight system. Hiller, who built the apparatus, first used it to make pure nitrogen bubbles. He anticipated that their properties would be similar to air, which is 80 percent nitrogen.

To our surprise, pure nitrogen bubbles made hardly any light. We therefore expected that oxygen would prove quite amazing. But again, we found that a pure oxygen bubble was very dim. Similarly, an 80–20 mixture of nitrogen and oxygen was a weak emitter. So was gas from a liquid-air canister! We anxiously searched for our stupid mistake.

In fact, the measurements were good. Air is 1 percent argon, and argon is removed from commercial liquid air. Adding argon back boosted the light intensity. Helium and xenon also worked, although each noble gas produces a unique spectrum. A small gas impurity of about 1 percent seems to be the key to sonoluminescence. We do not yet know why that is the optimal amount.

In view of our experimental results, what do we understand about sonoluminescence? First and foremost, we are dealing with a "virtuoso" sound field, one that positions a gas bubble at just the right location to act on it symmetrically and with maximum force. The theory of an adiabatic compression followed by an imploding shock wave provides an appealing picture that is helping to guide research.

Still, this working model must be viewed as tentative, because it fails to explain so many unknowns. These mysteries include the size of the bubble, the role of inert gases and the mechanism of light emission. Most important, theory and experiment have failed to determine the limit of energy focusing that can be achieved. Surely the mechanism is nature's most nonlinear system, yet it can be controlled and made free of chaos. The joy of this problem is that the effect is so robust but so sensitive that whenever we change a parameter we find wonderful new physics.

FURTHER READING

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