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Sonoluminescence: Sound science makes light work

A new phenomenon, in which tiny bubbles created in a liquid using ultrasound, will emit light, opens up an exciting "new window" in basic research. Mark Wilsey reports.

Tiny bubbles are more likely to bring to mind such things as a New Year's champagne toast, a soapy bath, or the fizz of seltzer tablets in water, than their role in exploring fundamental science. By the same token, while exposure to a loud noise can certainly jar one's nerves, there is a new field of technology—sonoluminescence—which is attempting to harness the energy from sound for various applications, from materials processing to water treatment. This phenomenon, in which light is produced from sound, is called sonoluminescence, in which a bubble is created in a liquid using ultrasound, and as the bubble collapses, it emits light.

Ultrasound (sound at frequencies above the range of human hearing) has long been used in industry for such applications as cleaning parts or mixing solutions. Ultrasound is familiar to many in medicine, where it is used to peer into an expectant mother's womb or to break up gall stones.

However, the sonoluminescence effects of ultrasound had gone largely unexplored.

Scientists have known of sonoluminescence for decades. In the mid-1930s, it was observed that photographic plates submerged in a solution could become fogged when exposed to an ultrasonic field. Later it was determined that the light emissions were coming from bubbles that had been formed by the acoustic waves.

The mechanical energy from the acoustic waves produces areas of high and low pressure in the liquid. Under certain conditions, the pressure is low enough to allow a small bubble to form from the dissolved gases in the liquid, through the process of cavitation. In cavitation, the acoustic wave pushes and pulls on the liquid; if the pull is great enough a small region of the liquid will be pulled apart to form a cavity, into which gases in the liquid can diffuse and produce a bubble. Then, as the wave pushes on the liquid, the pressure increases, and the bubble shrinks, thus compressing the trapped gas. Again, if the conditions are right, the gas in this collapsing bubble will give off a brief flash of light. This amounts to several orders of magnitude in energy concentration, to go from acoustical energy to light emission (**Figure 1**).

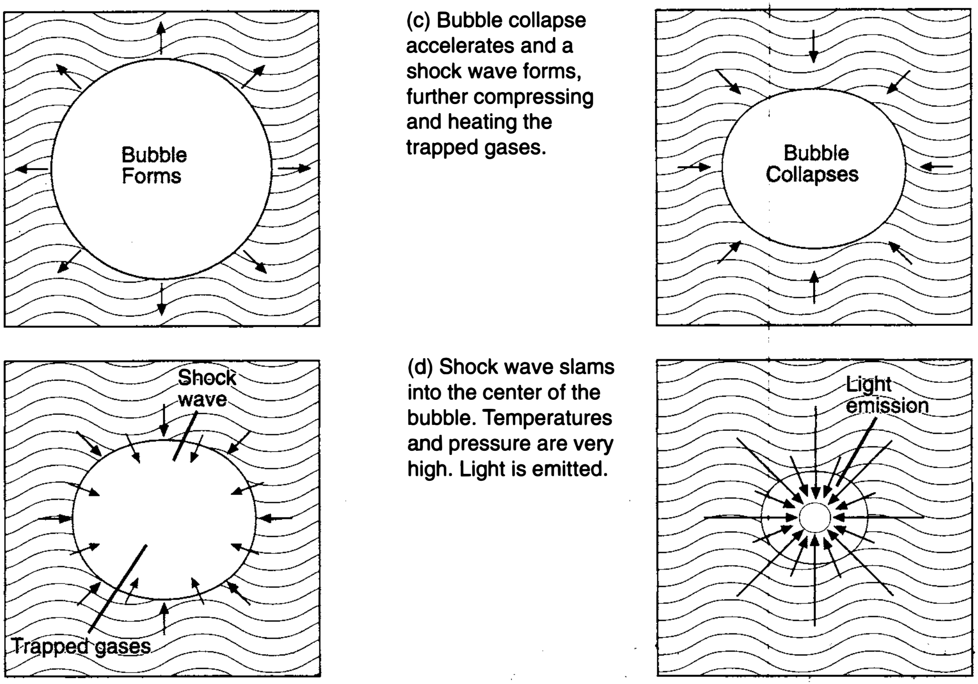
Around the turn of the century, cavitation was first explored in the field of ship propulsion. As the ship's propeller churns the water, it produces bubbles by cavitation. At higher speeds, some of the small bubbles cling to the propeller, thereby lowering its efficiency. But of greater concern was the fact that the bubbles' action eroded the propeller. Research into better designs and materials helped to alleviate the problem, but did not advance our understanding much.

Sonochemistry

Just as a propeller can produce a large number of bubbles, acoustic waves confined in a container can produce a lot of sonoluminescing bubbles. This multi-bubble sonoluminescence can be very useful. Kenneth Suslick of the University of Illinois at Champaign-Urbana, an expert on ultrasound, uses the multi-bubble effects of ultrasound to make new materials and affect the rates of chemical reactions. This field of study is known as sonochemistry. Here Suslick takes advantage of the unusual conditions inside the collapsing bubbles created by cavitation with ultrasound (**Figure 2**). The "hot spot" at the center of the bubble, a region just tens of microns (1 micron = 1 millionth of a meter) in size, can reach, by best estimates, temperatures of 5000 degrees Kelvin and pressures of 1700 atmospheres, for a very brief in-

FIGURE 1

Making light from sound



(a) Sound waves cause low pressure. Water molecules move apart, creating a bubble.

(b) Sound-wave pressure increases, the gases are compressed as the bubble collapses.

(c) Bubble collapse accelerates and a shock wave forms, further compressing and heating the trapped gases.

(d) Shock wave slams into the center of the bubble. Temperatures and pressure are very high. Light is emitted.

Source: *21st Century Science & Technology.*

stant. Each bubble is like a tiny chemical reactor, in which materials from a solution are brought together, heated and then cooled, or quenched, at an incredibly rapid rate, some 10 billion degrees per second. By comparison, a red-hot poker thrust into ice water would cool at a rate of a few thousand degrees per second.

Suslick has had great success in using this technique, with its high cooling rates, to produce amorphous metal compounds. These are materials that have been cooled so fast that they have solidified before crystallization can ocur. The resulting material has a porous, sponge-like structure, which is of interest for its possible properties as a catalyst. Catalysts are used to produce various chemical reactions with greater ease.

In a unique line of research, Suslick and his graduate student, Mike Wong, are developing methods of using ultrasound to synthesize microspheres of proteins. Such biomaterials could be used for more efficient delivery of medications or possibly as blood substitutes.

Single-bubble sonoluminescence

The multi-bubble effect has great advantages for the chemist who is interested in processing bulk materials. But for the physicist trying to study the phenomenon of sonoluminescence, the multiple bubbles make research more difficult.

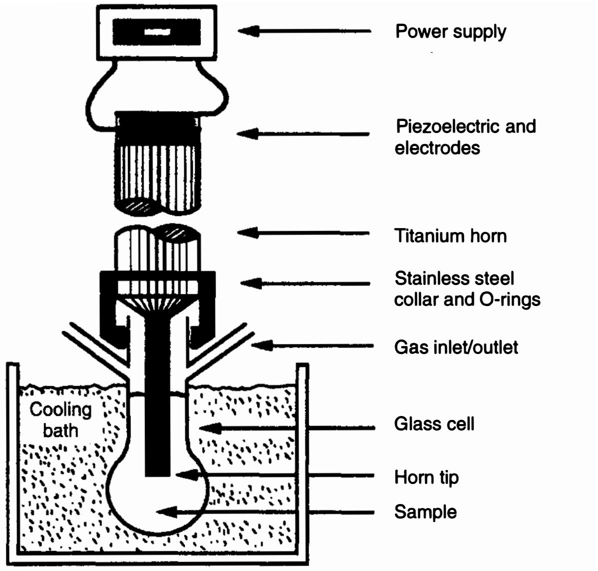
The breakthrough toward understanding the physics of sonoluminescence was made several years ago by researchers at the University of Mississippi, Lawrence Crum and his student Felipe Gaitan. Crum is now at the University of Washington in Seattle, and Gaitan is currently at NASA's Jet Propulsion Laboratory in Pasadena, California. Crum and Gaitan were able to produce sonoluminescence in an isolated, single bubble in the middle of a flask. Equally important, they could coax the phenomenon into producing a steady glow instead of just one brief flash. Now physicists had something that was much more accessible for probing.

The relative simplicity of the experimental setup was amazing. The basic components, which could be found in almost any university laboratory, consist of a spherical flask filled with water, a couple of transducers or loudspeakers, a frequency generator, and an amplifier. An electrical signal from the generator is sent to the amplifier which drives the transducers. The transducers, which are located opposite each other on the flask, convert the electrical signal into ultrasound. The flask acts as a resonator to intensify the sound, and at a specific frequency the sound is focused at the center of the flask. Here a single sonoluminescing bubble forms.

Since Crum and Gaitan's discovery, other laboratories

FIGURE 2

Sonochemical apparatus



Source: Kenneth Suslick.

have begun investigating sonoluminescence. Most notable is the University of California at Los Angeles, where a great deal of work has been done by Seth Putterman and graduate student Bradley Barber. They were the first researchers to attempt to measure the duration of the light emissions. Their work showed that the steadily glowing bubble was emitting light, or "flashing," with each cycle of the acoustic field, which is measured in microseconds (millionths of a second). However, the flash itself was found to be much shorter, on the order of picoseconds (millionths of a microsecond).

The researchers at UCLA found another remarkable aspect to these flashes. They found that the average interval between flashes in a 30-kilohertz (30,000 cycles per second) field was 33 microseconds, with a fluctuation of only 50 picoseconds over a period of 100,000 cycles, or just over 3 seconds. This greatly exceeded the rated stability of the frequency generator used. The precision of this periodicity could be illustrated as follows: Imagine a simple mechanical device, such as a watch, that is capable of striking a bell once a week and not varying the interval between rings by more than a second, week after week, for 2,000 years.

Work at UCLA has also shown that, while sonoluminescence in air bubbles is strong, bubbles of pure oxygen or nitrogen (the major components of atmospheric air) do not work nearly as well, and sometimes, they do not work at all. However, adding trace amounts of argon or xenon greatly improves the sonoluminescence of the nitrogen bubble.

Moreover, one of the limitations on the experiments is the brightness of the bubble: While it is bright enough to seen with the eye, it is still too dim for some of the instruments to measure. In order to overcome this, experiments with different liquids and gases are being performed to find out what effects they may have, with the goal of trying to produce a brighter bubble. Another approach has been to fiddle with the sound field, but here the system is also delicate and tolerances are low. There seems to be only a narrow range of acoustical amplitudes over which the bubble is stable. If it is too low, the bubble does not glow; if it is too high, the bubble disappears. Therefore, the problem becomes: How does one improve the experiment to more efficiently concentrate energy into the bubble?

Another narrow tolerance, demonstrated by Anthony Atchley at the Naval Postgraduate School in Monterey, Calif. and his colleagues, is that sonoluminescence is very sensitive to changes in the frequency of the sound field. Atchley et al. found that moving away from the resonance frequency at which the sonoluminescence is stable, even by a small amount, leads the bubble's emissions to become erratic or quasiperiodic.

Other measures of sonoluminescence

The work at the Naval Postgraduate School in studying sonoluminescence has been in three areas: measuring the optical spectrum of sonoluminescence, measuring the size of the bubble, and measuring the duration of the pulse. Their work confirms many of the findings of other laboratories.

In measuring the spectrum, they looked at different liquids for sonoluminesence, primarily water and mixtures of water and glycerin. The spectrum was found to be broad, increasing in intensity toward the ultraviolet.

The spectrum was measured by placing an optical fiber of quartz close to the bubble, in order to eliminate some of the light absorption caused by the water, which absorbs wavelengths shorter than about 210 nanometers (billionths of a meter). However, quartz starts to absorb at around 170 nanometers. In any case, there is still a little water present at the tip of fiber. To improve their measurements, the experimenters would like to go down to at least half that wavelength, 80 to 90 nanometers, well into the ultraviolet region of the spectrum.

In multi-bubble sonoluminescence, the temperatures inside collapsing bubbles reach several thousand degrees. The light emission is produced by highly excited states of molecular species like diatomic carbon, C_2 (a molecule made up of two atoms of carbon). However, in single-bubble sonoluminescence, the collapse of the bubble appears to be more efficient and generates much higher temperatures. The emission is not molecular but is likely some kind of plasma emission. This would come from atoms that have had electrons stripped away to become ionized.

Researchers at Los Alamos National Laboratory in New Mexico and at the California Institute of Technology are looking into the possibility of using sonoluminescence for waste water treatment. The temperatures inside the bubbles are high enough to cause compounds, such as solvents, in the water to break apart. The ultraviolet emissions from the bubbles could be effective in killing bacteria in the water.

Another area of research at the Naval Postgraduate School involved measuring the size of the bubble with a laser scattering technique. A laser is shone on the bubble, which scatters the light. Two detectors are used to look at the scattered light from two angles for greater accuracy. The

Federal funding falls to partisan posturing

On July 12, a senseless debate in the House of Representatives stripped research into sonoluminescence of its chance for federal funding. As a result, a tiny $1 million appropriation into the field was removed from the Department of Energy's (DOE) appropriations.

Earlier this year Rep. Dana Rohrabacher (R-Calif.), chairman of the Subcommittee on Energy and Environment of the House Committee on Science, commented during hearings on the sonoluminescence work at Lawrence Livermore National Laboratory, and tied it to the possibility of obtaining fusion energy. The Livermore researchers were quick to point out to Rohrabacher's office that sonoluminescence research is a basic science project, in an effort to dispel any notion of its foreseeable use as a source of fusion power.

Nonetheless, when the report on the Energy and Water Development Appropriations Bill, 1996, came out, the following was included under basic energy sciences for the DOE: "Within available funds, $1,000,000 is provided to fund peer-reviewed research on the potential energy applications of sonoluminescence." This money was clearly earmarked for Livermore.

One million dollars would be a drop in the bucket of DOE's overall budget, but would be a major boost to sonoluminescence research, which has heretofore operated on a shoestring. With a $1 million budget, experiments could be carried out that would begin to bring our understanding of the phenomenon to a new level.

Rohrabacher's office tried to justify the funds as being a small investment with a potentially large return, but even that time-tested battle cry was not enough to rally House Republicans to stop an amendment by Rep. Mike Ward (D-Ky.) to strike them. To his credit, in introducing his bill, Ward conceded that sonoluminescence was a "legitimate course of study," and that the funding was not "a piece of pork."

So what's his beef? Since neither the DOE nor Livermore requested the money, it is then specially earmarked. And Ward and many other congressmen are against earmarks in general, which are usually a means of funding pet projects. But what really stuck in his craw was that there was not any mention of this money in any hearings, and that it was part of some 60 pages of report language which was added to the bill. According to Ward's office, the subcommittee's Democrats argued that they had not had sufficient time to review the material in the report and therefore opposed its inclusion, but they were outvoted by the Republicians.

Responding to Ward's bill to cut these funds, Rohrabacher argued, "This is exactly the kind of program the federal government should be doing." He continued that "small research programs that have high potential . . . never get the money, because they do not have lobbyists." But Rohrabacher is not without his axe to grind, i.e., to support the small programs and to chop away at "megaprograms," a pragmatic approach that ignores their complementarity.

In a arrogant display of smug sarcasm, Rep. Fortney Stark (D-Calif.), whose district includes Livermore, blasted the research: "This is a wonderful project," he said, "shooting light on these bubbles will cause a lot of wonderful things." Then he continued, "Do you know what else they make in Livermore, California? . . . It is right in the middle of the finest champagne country in the world. What this will do is irradiate that champagne that comes from California, much to the disadvantage of New York, where they do not make such very good champagne. . . . I want to say to you that if you want to waste $1 million trying to make California champagne better, which you cannot do, then we welcome this money."

Stark's theatrics had the desired affect. When the House vote was taken, Ward's bill passed 276-141. Some 85 Republicans joined in voting against it.

While such bipartisan short-sighted, know-nothing pragmatism should not be surprising, it makes one wonder: If a small, creative science project with such great potential for broadening our knowledge of the physical universe, can be cut, what's left?

amount of scattering is then used to determine the bubble size. By using a pulsed laser, the bubble is examined at various parts of its cycle.

The researchers were able to measure the bubble's size to within 10 nanoseconds of its final collapse and emission. At this time, the bubble is still a few microns in radius. At times closer to emission, the bubble is so small that its diameter is approaching the wavelength of the laser light and the scattered signal becomes very weak.

The third area concerned measuring the duration of the pulse. Researchers at the Naval Postgraduate School asked Michael Moran at Lawrence Livermore National Laboratory in California, for help in measuring the optical spectrum of sonoluminescence and the time-dependence of the emissions. Yet, even with the ultrafast cameras available at Livermore, the duration of the pulse could not be definitively measured. The results indicated that the pulse was very fast, possibly faster than the 50 picoseconds that other researchers had indicated.

Research at Livermore

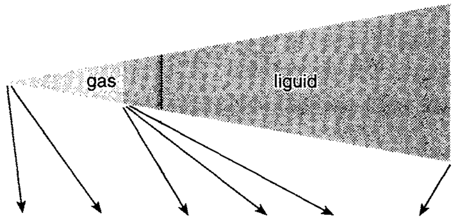
The collaboration with Livermore was a natural teaming of researchers and resources. Sonoluminescence is a type of phenomenon in which the skills and talents that a laboratory, such as Livermore, has developed over decades in nuclear weapons and fusion research can very effectively be brought to bear. Sonoluminescence research at Livermore has taken on something of a life of its own.

Analogies have been made that compare sonoluminescence to inertial confinment fusion (ICF). In ICF, a powerful energy source, either lasers, X-rays or particle beams, is directed onto a small, spherical target containing fusion fuel. The energy vaporizes the outer shell of the target, and the reactive force of the expanding plasma compresses the fuel inside the target. If the compression is stable enough, and the temperature and pressure are high enough, a burst of fusion energy is produced. Livermore is the home of NOVA, one of the world's largest lasers for conducting ICF research. The question then arises: How are the physical conditions of a collapsing bubble and a exploding fusion target similar? This is the point that Livermore takes up.

Livermore physicist William Moss has carried out some of the most sophisticated calculations to date on bubble growth and collapse in single-bubble sonoluminescence (**Figure 3**). He has performed the first fully compressible non-linear hydrodynamic computer simulations, which means that, in modeling this system, the compressibility of water is attempted to be taken into account. Typically, other models use the simplifying assumption of regarding water as incompressible, primarily because of the computational limits of the program and the computers being used. Moss says that these models are good for a first approximation, but to get a better understanding, a program needs to have included better physics.

FIGURE 3

Enormous range of temperature, pressure, and density, at time of 'flash'



Radius (µgm)	0.001	0.003	0.6	5	500	30,000
Temperature (K)	2.0M	0.6M	700	300	290	290
Density (g/c^3)	6.2	14.1	2.0	1.2	1.0	1.0
Pressure (bar)	43M	32M	0.3M	7,000	5	1.0

The chart depicts the conditions of temperature, pressure, and density at the time of the flash, for various distances in the liquid and bubble. The values are derived from a computer simulation by William Moss at Livermore. This is what his theory predicts, but it is still up to the experimentalists to record what is actually happening. Note that, at the interface of the bubble and the liquid, the density goes from 1 gram per cubic centimeter to 2 g/C^3, due to the compessibility of water, which Moss's model take into account.

Source: William Moss/Lawrence Livermore National Laboratory.

Moss starts with equations for the state of the gas, and incorporates the liquid, and even the glass flask, in an effort to simulate as much of the problem as possible. He began with a standard hydrodynamic program, but is now using some of the weapons computer codes, "which are optimized a little better for these kinds of problems," Moss says.

It should be noted that there is, as yet, no direct experimental measurement of temperature and pressure in single-bubble sonoluminescence. The temperatures and pressures that have been reported by various researchers are, for the most part, inferred from the spectrum or are based on theoretical calculations.

Nonetheless, what Moss's model shows, is that, when a bubble contracts, a shock wave is generated that slams into the center of the bubble. The bubble radius, in Moss's calculations, is about half a micron. It is only its inner portion that becomes really hot—between 100,000 and 1 million degrees, according to his calculations—with pressures reaching 100 million atmospheres. These conditions last for about 10 picoseconds, as the shock wave crashes in and then bounces out.

Moss believes that the rapid bounce is caused by the intermolecular potential: That is, as the atoms are pushed closer together, the repulsive force between them becomes stronger, which also causes the short time-scale. "What we're talking about," Moss explains, "is just the mechanical process."

Moss notes that one of the factors controlling temperature is ionization. Ionization of a material removes electrons, thus removing energy that would otherwise go into temperature; therefore, something that becomes significantly ionized, will not become as hot as something that does not become as ionized.

It was this realization that led Moss and Moran to look at deuterium, an isotope of hydrogen which is one of the fuels used in fusion. Deuterium, like hydrogen, has only one electron. Moss's team believed that, once the electron was removed, the energy used to compress the deuterium bubble could then go into raising the temperature, thereby making the deuterium very hot compared to air. However, it turns out that deuterium is much more compressible than air is at high densities, so it is harder to get it as hot, given the same experimental conditions. The calculations indicate that the air will get hotter, because it has a stiffer repulsion potential.

Moss believes that one way to overcome this limitation is to shape the pulse of the driver, the acoustical wave. So far, the acoustical wave that researchers have used is a simple sine wave, evenly rising and falling in pressure over time. By changing that pulse, by putting a well-timed spike of positive pressure on top of the oscillatory field, one could "give the bubble a real good whomp at just the right time," says Moss. His calculations have shown that such an approach would have a large effect, but this has yet to be borne out experimentally.

This research approach is what led them to start working on a deuterium experiment and to look at the "remote possibility" of producing the conditions for a fusion reaction within the sonoluminescing bubble, Moss said. In comparing sonoluminescence to inertial confinement fusion, there are some similarities, but also big differences. For one thing, there is an enormous amount of material in a fusion target as compared to a sonoluminescing bubble, and the temperatures and densities are also much higher in the fusion target. But the important difference, Moss is quick to point out, is that ICF works: It produces fusion. However, Moss added, "I would say, to be as optimistic as we can and to be scientifically honest, that what we think we've shown is, that it may not be impossible to get fusion this way."

Although Moss is hopeful, he said that even if fusion does not occur, there should be a wealth of physics coming out of research into sonoluminescence. "It seems to me that you can't lose scientifically," Moss said.

On a broader point, Anthony Atchley at the Naval Postgraduate School, notes that sonoluminescence puts in the experimenter's hands, at a very economical price, a new area to study, a region that in general has not been studied before. "It is a different window on the world," he said, "and, who knows what you could find with that?"