Sonoluminescence

Lawrence A. Crum and Ronald A. Roy

When trapped in sufficiently intense acoustic fields, single bubbles of gas can emit luminescence bright enough to be visible in an undarkened room. The large number of intriguing results recently published about such single-bubble sonoluminescence (SBSL) (*1–9*) suggests that this phenomenon awaits a full explanation. And as reported by Hiller *et al.* on page 248 of this issue (*10*), some exciting atomic physics may be occurring within the collapsing cavitation bubble that gives rise to SBSL. However, many of the results they present are also anomalous and defy immediate explanation.

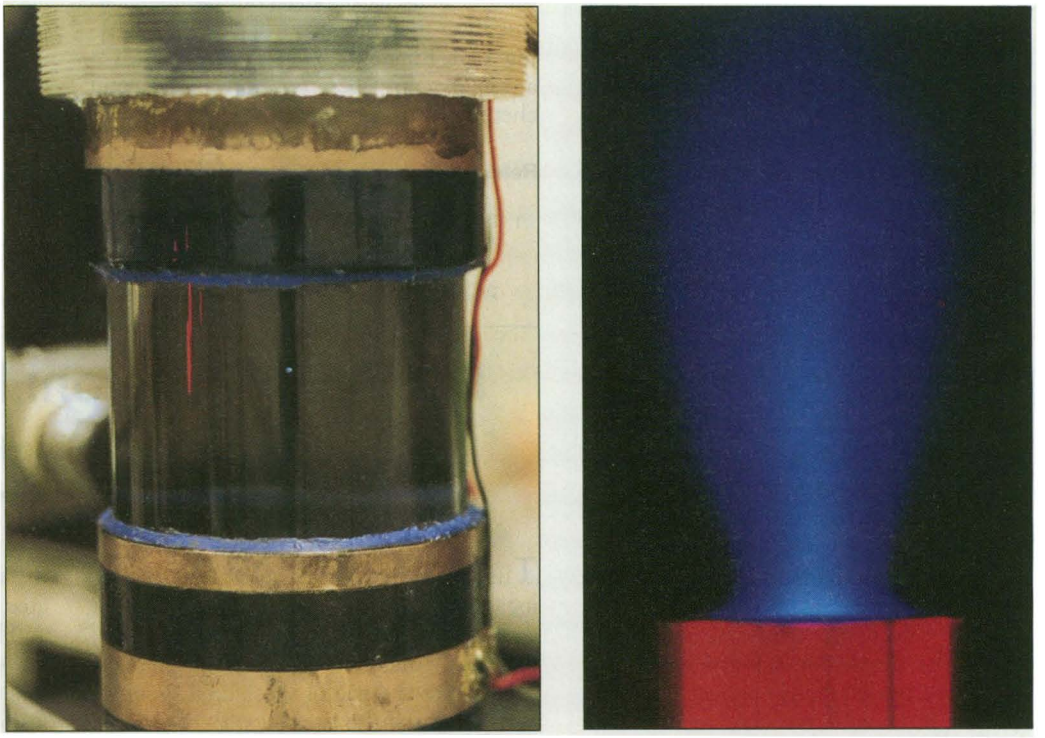
Sonoluminescence (SL) was discovered nearly 60 years ago when Frenzel and Schultes (*11*) found that photographic plates became exposed when submerged in water and irradiated with ultrasonic waves. Since then, the phenomenon of SL has been associated with the presence of (relatively) intense sound fields within liquids. The light emission implies existence of high local temperatures (the ambient temperature of the liquid remains relatively constant), temperatures high enough to incandesce gas and influence chemical reactions. Thus arose sonochemistry, which has resulted in an active field of both basic research and emerging technology development (*12*).

When an intense sound field is produced within a liquid, microscopic cavities of gas or vapor can be generated when the liquid fails under tensile stress. The subsequent acoustic compression cycle forces these cavities to collapse violently, which results in a remarkable concentration of mechanical energy, estimated to be as high as 12 orders of magnitude (*3, 4, 10*). In the process, the gas contained within these cavities is heated to luminescence. In some cases, the light emissions appear to come from a distributed region surrounding the tip of the acoustic source. This multiple-bubble sonoluminescence (MBSL) results from the collapse of many separate, individual cavitation bubbles. Because MBSL occurs randomly and transiently, it has been difficult to study in much detail. Recently, Gaitan and co-workers discovered the unique conditions necessary for a single bubble, pulsating stably, to emit SBSL each acoustic cycle (*1, 2*) (see figure); its robust stability and simplicity has permitted detailed studies of this "hydrogen atom of sonoluminescence" (*1–4, 10*).

Theoretical calculations of the time interval during which the contents of a gas bubble, driven to collapse by the sound field, is sufficiently heated to emit light tend to be on the order of tens of nanoseconds. However, when Barber and Putterman sought to measure the duration of the light flash, they discovered that they were in fact measuring the impulse response of their photomultiplier tube (PMT) (*3*). Even subsequent measurements obtained with the world's fastest multichannel plate PMT led to results that were rise time–limited. Their best measurements to date indicate an upper bound of about 50 ps, about 1/1000 of that predicted by conventional theory of cavitation bubble collapse. In addition, Barber and Putterman measured the statistics of the flash-to-flash interval for a bubble driven at 30 kHz and discovered that it had an average value of 33 μs with a root-mean-square fluctuation of only 50 ps (*3*). This remarkable stability, 1 part in 10^6, exceeds the rated stability of the frequency generator that drove the acoustic system.

The spectrum of MBSL contains molecular emission bands associated with the liquid in which the sonoluminescence occurs (*12*). For example, if MBSL occurs in an organic liquid such as dodecane, one sees diatomic carbon bands; if MBSL occurs in an inorganic liquid such as water, one observes the spectral bands of the OH radical; these bands suggested temperatures within the cavities on the order of 5000 K. However, when Hiller *et al.* (*4*) and Atchley and coworkers (*5*) studied SBSL emissions, they discovered a smooth spectrum, devoid of any distinct bands. This suggests the presence of much higher temperatures, perhaps orders of magnitude greater than those encountered in MBSL systems.

To date, only one viable explanation has been offered for the short pulse duration of the SBSL flash and for the seemingly extreme temperatures involved: a



Bright bubbles. An acoustic standing wave levitates a small gas bubble near the center of a glass cell **(left)** and drives that bubble to radial excursions of sufficient amplitude to generate sonoluminescence each and every acoustic cycle. Note the bright spot at the center of the cell, which can easily be seen without darkening the room; no chemical enhancement is required. This bubble was driven at a frequency of 22.3 kHz and at a pressure amplitude of about 1.3 bar. In an example of MBSL **(right)**, the intense sound field near the tip of an acoustic source produces many transient cavitation bubbles that grow and collapse with such violence that they heat their respective interiors to incandescence. Because the individual bubbles persist for only a few acoustic cycles (at 22 kHz), they are not visible in this photograph. Luminol, a wavelength shifter, was added to enhance the light emission, which is normally too faint for unaided photography.

The authors are in the Department of Acoustics and Electromagnetics, Applied Physics Laboratory, University of Washington, Seattle, WA 98105, USA.

spherically converging shock wave generated within the collapsing bubble. Wu and Roberts (*7*) as well as Greenspan and Nadim (*8*) have demonstrated numerically that such an imploding shock should exist in the SBSL bubble and that extremely short pulse durations (0.1 ps) and high temperatures (1000 eV; 1 eV = 11,600 K) should occur. Using a more accurate equation of state, Moss *et al.* (*9*) have confirmed predictions of extreme temperatures and pressures, obtaining values more in line with the (crude, at this time) experimental measurements, namely, pulse durations on the order of 10 ps and peak temperatures on the order of 10 to 100 eV. Furthermore, their computations suggest that at various locations within the imploded core at the center of the bubble, pressures can be as high as 200 Mbar (1 Mbar = 10^{11} Pa), and densities as high as 13.4 g/cm^3 (at these levels, it is possible that the compressed air near the center of the bubble will have properties similar to that of a metal). Note that all the calculated results cited above are based on one-dimensional calculations assuming a perfectly symmetrical bubble collapse and are mitigated by the effects of dissociation and ionization [which are accounted for in the Moss *et al.* (*9*) computations] and by various radiation and mass transport mechanisms (which are not accounted for by Moss *et al.*).

As indicated in figure 2 of Hiller *et al.* (*10*), a small quantity of argon introduced into a pure nitrogen bubble increases the luminosity of SBSL by nearly two orders of magnitude. What effect does argon have on this system? Does it strongly influence the dissociation, ionization, and radiation transport within the core? Does it have a catalytic effect on electronic transitions within the plasma or material composing the core? Does it readily conduct heat from the hot interior of the core to the outer layers and thus increase the radiated energy or the total volume of high-temperature gas? These questions are difficult to answer with the existing data and clearly require additional measurements and computations.

Figure 3 of Hiller *et al.* (*10*) demonstrates that if the bubble contains certain gas species, the spectra show broad peaks near 300 nm, whereas for other species, no peaks exist and the spectrum monotonically increases down to the water cutoff (the transmissivity of the ultraviolet through water is greatly reduced below 200 nm). Why is it that a maximum exists at all? If the gas core is heated and compressed to the degree predicted by recent theories, then only the outer shell should radiate (like the sun). If there is a broad maxima for xenon, then shouldn't there also be one for helium? These again are anomalous results and perhaps have something to do with the heat transport through the compressed gas.

Finally, the data displayed in figure 5 either have a trivial explanation (for example, a periodic detuning of the cell) or they are truly remarkable. These data suggest that some mechanism, possibly gas diffusion across the gas-liquid interface, is causing the luminosity and equilibrium bubble radius to cycle with a period on the order of seconds. It seems remarkable to us that such long-term memory (on the order of 100,000 acoustic cycles) could exist in a mechanical system.

As we currently understand it, single-bubble sonoluminescence may result in temperatures in excess of 10^5 K, pressures in excess of 10^7 bar, light emissions lasting less than 50 ps, and mechanical energy concentrations of up to 12 orders of magnitude; all this from a simple acoustical system costing a few hundred dollars to construct. It is a remarkable laboratory for physics and chemistry.

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Flu Virus Invasion: Halfway There

Chavela M. Carr and Peter S. Kim

The protective barrier provided by biological membranes is exceedingly difficult to breach. The fusion of two distinct lipid bilayers is therefore energetically unfavorable in the absence of specialized proteins. Fertilization of the egg cell with the sperm, perhaps the most dramatic consequence of membrane fusion, requires the sperm protein PH-30α-β (*1*). Enveloped viruses also use membrane fusion: Infection of animal cells by human immunodeficiency virus (HIV), for example, is aided by the HIV envelope protein gp120-gp41 (*2*).

The best characterized membrane fusion protein is the hemagglutinin (HA) protein found on the surface of influenza (flu) virus. In spite of decades of research, the mechanism by which HA induces fusion remains elusive. Recent developments, however, including a report by Shin and co-workers on page 274 of this issue of *Science* (*3*), suggest that we are getting closer to an understanding of this protein-mediated membrane fusion process.

Flu onset begins with the binding of influenza virus to nasal epithelial cells. In a function that is distinct from membrane fusion, HA mediates viral attachment to sugar groups (sialic acid) on the surface of the host cell. Binding does not lead directly to membrane fusion, since HA in its native state is not fusion active. Instead, the cell internalizes the bound virus and surrounds it with an endosome. Only in the mildly acidic conditions of the mature endosome does the HA protein switch from an inactive to a fusion-active (fusogenic) state. In this fusogenic conformation, HA promotes fusion of the viral and cellular membranes, leading to release of the nucleocapsid into the cytoplasm and thereby initiating replication and proliferation of the virus.

But how does HA induce membrane fusion? Thirteen years ago, the x-ray crystal structure of HA in the native state was reported by Wiley and his co-workers (*4*). Hemagglutinin is a trimeric protein with three identical subunits that span the viral mem-

The authors are in the Howard Hughes Medical Institute, Whitehead Institute for Biomedical Research, Department of Biology, Massachusetts Institute of Technology, Cambridge, MA 02142, USA.

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