Bubble Puzzles

Bubbles are familiar from daily life and occupy an important role in physics, chemistry, medicine, and technology. Nevertheless, their behavior is often surprising and unexpected—and, in many cases, still not understood.

Detlef Lohse

With their ubiquitous occurrence in a multitude of fluid systems, bubbles occupy an important place in contemporary science and technology. One can readily cite several examples: the production and transport of oil, in which bubbles are purposely injected to help lift heavy oil to the surface; energy generation, in which boiling is the key process in producing the steam to drive turbines; the chemical industry, in which gas–liquid reactors rely on bubbles to increase the contact area between the phases; the oceans, in which bubbles generated by breaking waves are important sinks for atmospheric carbon dioxide; piezoelectric ink-jet printing, in which they are just disturbing; and bubble chambers in high-energy physics, in which they used to signal the traces of energetic particles.

Bubbles are also fascinating in their own right, from the most innocent-looking problem—a rising bubble in still water—to their formation, oscillation, and collapse. A bubble's collapse can be extremely violent, as revealed in light emission, called sonoluminescence. Some shrimp use the violence of the collapse to kill prey, and many technological applications such as ultrasound cleaning and sonochemistry also utilize it.

Rising bubbles

The simplest building block of bubble systems is a single gas bubble in still water. One expects that it rises straight upward, due to the buoyancy force that is directed opposite gravity. However, bubbles with a radius larger than about 0.8 mm spiral or zigzag as they rise. Why? Leonardo da Vinci first pointed out this phenomenon and even drew rising spiraling bubbles.[1] The question has now been tackled for decades, and although the phenomenon is ubiquitous in nature, technology, and even popular toys such as bubble columns, the full answer is not yet known. The difficulties arise from the bubble's interaction with its own wake, from the free and thus deformable surface, and from surface impurities that are unavoidable even in ultraclean water.

For bubbles in turbulence or for many interacting bubbles, the question is even more difficult to answer. Accurately calculating the dynamics of a few air bubbles in turbulent flow is numerically still infeasible. Approximations are therefore required. One approximation is to replace the sum of all stresses over the moving bubble–liquid interface by effective size-dependent forces[2] such as the drag force, lift force, added-mass force, and so-called history force (which is nonlocal in time) and to approximate the effect of the bubble on the flow by a point force. For larger bubbles, all the approximations naturally get worse; in that regime, the bubble's shape also shows strong deviations from sphericity, as described in the box on page 37.

Detlef Lohse *(lohse@tn.utwente.nl) is chair of the physics of fluids group in the department of applied physics at the University of Twente in Enschede, the Netherlands.*

Bubble formation

Bubbles can be injected in some fluids, but they can also form spontaneously. Such spontaneously formed bubbles mainly contain liquid vapor instead of some other gas. This process of bubble formation, familiar to all of us from boiling water, is called cavitation (or, more precisely, nucleation).[3] Cavitation can occur in a liquid when the local pressure $p(\mathbf{x})$ drops below the vapor pressure p_V of the fluid (see the article by Humphrey Maris and Sebastien Balibar, PHYSICS TODAY, February 2000, page 29). One way to achieve cavitation is to increase the liquid's temperature, because the vapor pressure is temperature dependent: For water at 20°C, the vapor pressure is 0.023 bar (2.3 kPa), but at 100°C, it is 1 bar, and thus the water boils.

Another way to achieve cavitation is to increase the local flow velocity $U(\mathbf{x})$. An easy experiment is to reduce the cross section of a pipe in one region, making a so-called diffuser that produces large local flow velocities due to mass flux conservation. For steady potential flow, the corresponding local pressure $p(\mathbf{x})$ can be estimated from Bernoulli's equation,

$$p(\mathbf{x}) + \frac{1}{2}\rho U^2(\mathbf{x}) = \text{constant}. \tag{1}$$

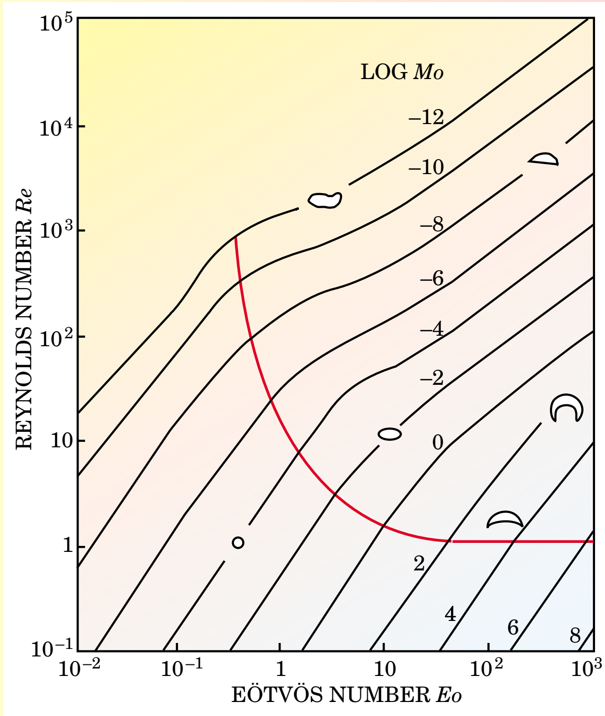
At an ambient reference pressure of 1 bar and at room temperature, a water velocity of about 14 m/s is sufficient to nucleate bubbles.

Bernoulli's estimate does not consider viscous effects, the gas content of the fluid, impurities, or walls and other inhomogeneities. Indeed, in extremely purified water, cavitation occurs at much larger tensions ("negative pressures") than in normal water—but still far from the value calculated from the attractive van der Waals forces between the water molecules. Crevices at surfaces or remaining impurities to which submicron gas bubbles attach seem to play a prominent role in the bubble nucleation process, but our understanding of cavitation is still incomplete.

Oscillating bubbles and the sound of rain

What happens to gas bubbles when the pressure is oscillating periodically? Due to the gas compressibility, the bub-

Dimensionless Numbers for a Rising Bubble



In fluid dynamics, the absolute values for the system size, the flow velocity, or the fluid density are not important for the behavior of the system. Because the underlying dynamical equations can be rescaled, all that matters are the ratios of parameters, such as the ratio of inertial forces to viscous forces or of capillary forces to gravitational ones. In other words, what is crucial are the dimensionless numbers.

For a single rising gas bubble in still fluid, the physical quantities are the bubble volume V [or the equivalent spherical diameter $d = (6V/\pi)^{1/3}$], the fluid kinematic viscosity ν, the surface tension σ, the density ρ, and the gravitational acceleration g. For water at 20°C, these material constants are $\nu = 10^{-6}$ m^2/s, $\sigma = 0.072$ kg/s^2, and $\rho = 1000$ kg/m^3. The two independent parameters of the system are the Eötvös number

$$Eo = \frac{\rho d^2 g}{\sigma},$$

which is the ratio of the gravitational and the capillary forces, and the Morton number

$$Mo = \frac{g\nu^4\rho^3}{\sigma^3},$$

which only depends on fluid properties. Water at 20°C has $Mo = 3 \times 10^{-11}$.

The system responds with some rise velocity $U(Eo,Mo)$, which can be made dimensionless in many ways: as the Reynolds number $Re = Ud/\nu$, comparing inertial and viscous forces; as the Weber number $We = \rho U^2 d/\sigma$, comparing inertial and capillary forces; or as the drag coefficient $C_d = 4gd/3U^2$, comparing gravity and inertial forces. Various experimental, theoretical, and numerical studies have been conducted to determine $Re(Mo,Eo)$, mainly for air bubbles in water,[2] but a rigorous understanding could only be achieved in limiting cases.

A sketch of the parameter space of rising bubbles is shown in the figure. The thin lines characterize constant Morton number. The bubble shapes in the various regimes are sketched; the thick red line indicates the transition toward nonspherical bubbles. (Figure adapted from R. Clift, J. R. Grace, and M. E. Weber, *Bubbles, Drops and Particles*, Academic Press, New York, 1978).

ble also will oscillate periodically around the ambient radius R_0 that the bubble would have under static, ambient conditions.

If instead the bubble is kicked with a single pressure pulse, the bubble's resonance frequency f_0 survives longest; all other frequencies damp out earlier. To calculate the resonance frequency, one needs the restoring force, which results from the pressure in the gas bubble. For large enough bubbles, $R_0 \gg \sigma/P_0 \approx 1\ \mu m$, the force depends on the ambient pressure P_0 and the actual radius $R(t)$, and the resonance frequency is given by[3]

$$f_0 = \frac{1}{2\pi}\sqrt{\frac{3\gamma P_0}{\rho R_0^2}}. \qquad (2)$$

Here γ is the adiabatic exponent, the ratio of the constant-pressure and constant-volume heat capacities of the gas. For air bubbles (for which $\gamma = 1.4$) in water under standard conditions, equation 2 reduces to $f_0 R_0 \approx 3$ kHz mm.

Most remarkably, this eigenfrequency of bubbles can be heard as the underwater sound of rain. When raindrops fall on a calm lake, the underwater sound is *not* generated at drop impact. Rather, at impact, a small air bubble is entrained, as shown in figure 1. Due to the violent entrainment process, the bubble experiences a pressure kick and subsequently oscillates at its eigenfrequency.[4] We hear the corresponding sound emission from the resulting pressure field

$$P_s(r,t') = \frac{\rho R}{r}\left(2\dot{R}^2 + R\ddot{R}\right) \qquad (3)$$

at large distances r from the bubble at the delayed time $t' = t + r/c$, where c is the speed of sound in water. Typically, the entrained bubble has a radius of about 0.2 mm, corresponding to a resonance frequency around $f =$ 15 kHz, which is in the audible range. If the raindrop is too small or too large, no bubble is entrained and the sound is shut off. Correspondingly, surfactants can suppress air entrainment and the sound of rain.[4]

Another example of air entrainment in liquid can be demonstrated in the following simple experiment.[5] When a cylinder partly filled with a water–glycerol mixture is vertically shaken close to the resonance frequency of the liquid column, a radial surface wave is generated. Due to a parametric (Faraday) instability, the radial surface wave steepens more and more. Eventually, the elevated fluid mass in the center of the cylinder has enough kinetic energy to create a cavity when falling down, as shown in figure 2. The cavity is then pinched off due to the hydrostatic pressure, and a big air bubble forms. At pinch-off, two fluid jets form, one upward that eventually shoots fluid into the air, and one downward into the entrained bubble. (See also the cover of this issue.) Moreover, at pinch-off, a second, much smaller bubble is formed (first

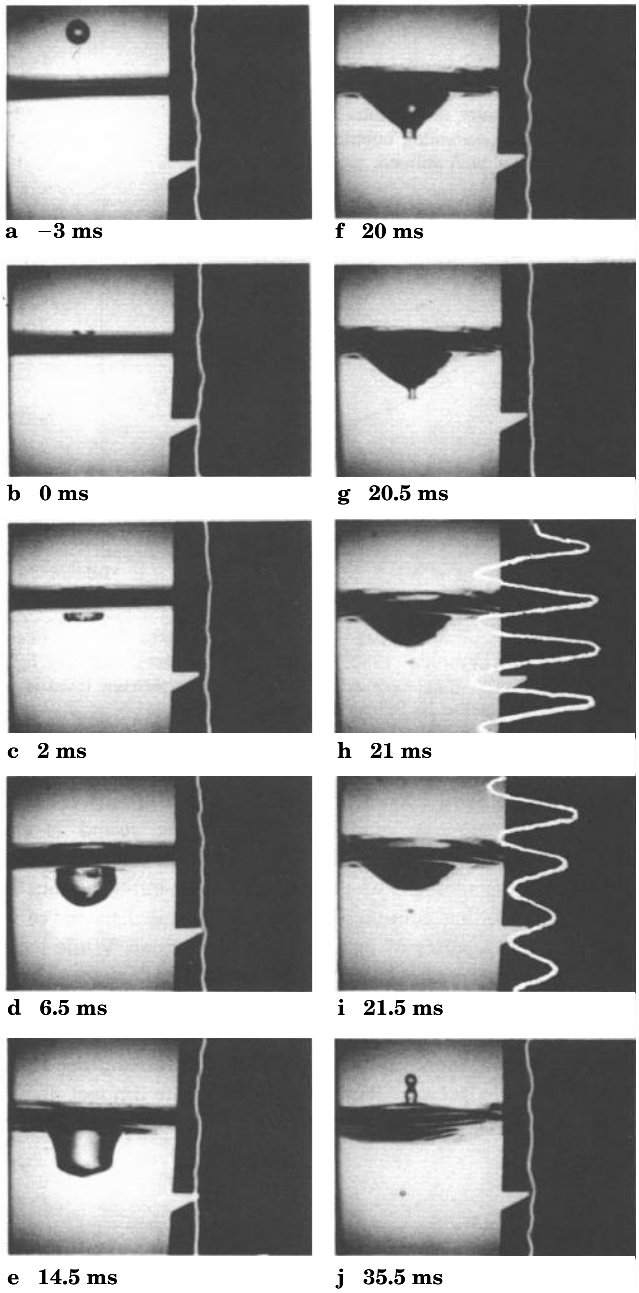


Figure 1. A drop falling on a water surface can create a bubble whose oscillations are audible as the sound of rain. Frames **(a)**–**(j)** show the progression of the bubble evolution; the emitted sound is plotted on the right in each frame. The drop diameter is 3 mm, and its velocity at impact is 2 m/s. No sound or bubble is produced at drop impact (b). The bubble first appears in (g), just as sound emission sets in. (From A. Prosperetti, L. A. Crum, H. C. Pumphrey, *Journal of Geophysical Research* **94**, 3255, 1989.)

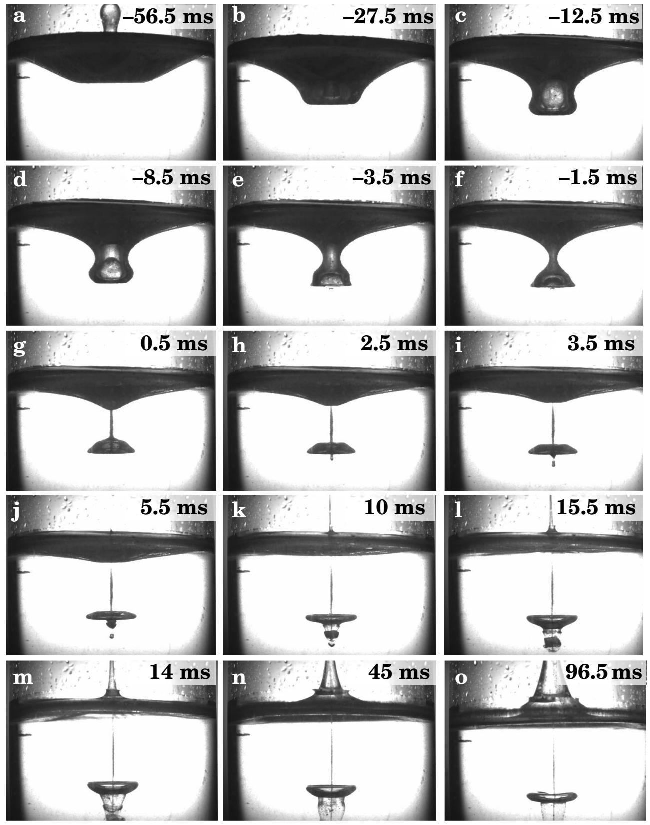


Figure 2. Air entrainment in an 11-cm-diameter cylinder partially filled with a water–glycerol mixture and shaken at resonance frequency (8.1 Hz).[5] The air–liquid interface is dark in these pictures due to the light reflection. After many oscillations, the elevated fluid mass in the center of the cylinder has enough kinetic energy to form a void when falling down **(a–d)**. The cavity is then pinched off **(e–g)**, leaving behind a stream of microbubbles. Upward and downward jets develop **(h–i)**. The upward jet eventually shoots fluid into the air, and the downward jet pierces the big cavity **(j–o)**. (Movie taken by Marijn Sandtke, physics of fluids group, University of Twente.)

visible in figure 2i) that then gets pushed away after the downward jet has completely pierced the big cavity.

Collapsing bubbles

Bubbles can also sound less melodic than the sound of rain. In World War I, the Royal Navy approached John William Strutt (Lord Rayleigh) with the problem of damaged propellers of fast-going boats and submarines. Rayleigh confirmed earlier speculation that collapsing bubbles were the origin of the damage. The phenomenon is now called cavitation damage: The ship propellers are rotating so fast that the pressure near the blades drops below the water vapor pressure. Consequently, bubbles form (see figure 3). The subsequent collapse can be so violent that the ship propeller gets damaged. Rayleigh mathematically described the dynamics[6] of such a collapsing void in water, assumed to be spherical with radius $R(t)$, and laid the foundation of what is now called the Rayleigh–Plesset equation,[3,7]

$$R\ddot{R}+\frac{3}{2}\dot{R}^2=\frac{1}{\rho}\left[p_g-P_0-P(t)\right]-4\nu\frac{\dot{R}}{R}-\frac{2\sigma}{\rho R}. \qquad (4)$$

Here ν denotes the kinematic viscosity, p_g the gas pressure inside the bubble (dependent on the radius), and $P(t)$ the time-dependent external pressure.

Rayleigh–Plesset dynamics can lead to energy focusing, as can be seen by neglecting all terms on the right-hand side of equation 4, that is, by considering only the inertial terms, $R\ddot{R} + 3/2\,\dot{R}^2 = 0$. Integration immediately gives $R(t) = R_0[(t_* - t)/t_*]^{2/5}$, with the remarkable feature of

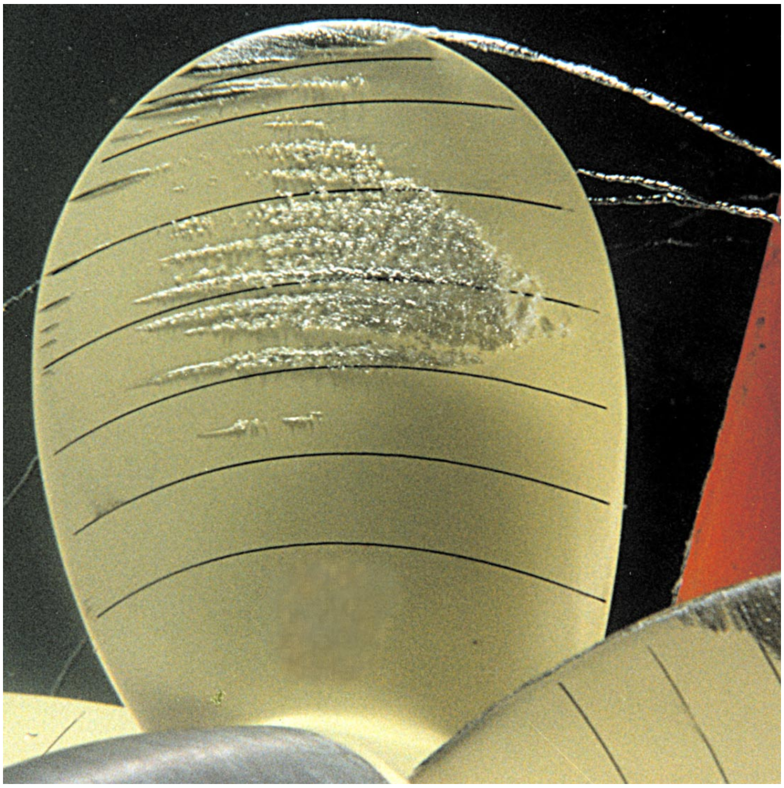


Figure 3. Cavitating bubbles generated by a fast ship propeller. Such bubbles can damage the propeller when they collapse next to it. (Photo by G. Kuiper, Marin, the Netherlands.)

a divergent bubble-wall velocity as t approaches the time t_* of the bubble collapse. It is this finite-time singularity that leads to the cavitation damage. The collapse is eventually cut off by the adiabatic compression (and thus heating) of the gas inside the bubble and by the sound emission at bubble collapse,[8] or in many cases also by the disintegration of the bubble. The emitted sound pressure (equation 3) obviously also diverges.

Even nowadays, cavitation damage to ship propellers is a limiting factor for the speed of boats. Due to Bernoulli's law (equation 1), cavitation is unavoidable at high speeds. So the art is to design the propellers so that the collapses occur away from the propeller and do not cause any damage.

However, cavitation damage can also be beneficial—at least for some species. So-called snapping shrimp (*Alpheus heterochaelis* and other species) use it to stun and even kill prey. The most distinct feature of these shrimp is a giant claw that can be closed rapidly and makes a loud popping sound. It was believed that sound originated from the claw halves' hitting each other. High-speed video imaging and parallel sound detection revealed that a thin water jet originates at claw closure.[9] The jet is so fast that, according to Bernoulli's law, a cavitation bubble develops, as shown in figure 4. When the bubble collapses, sound is emitted in the form of a shock wave—with fatal effect on the prey. The victim is then picked up by a second, normal-sized claw and eaten. The sound generated at bubble collapse has a broadband spectrum corresponding to the very narrow peak in time over which the bubble collapses. Because the snapping shrimp live in large colonies (in California's San Diego Bay or around Florida, for example), they can generate noise so loud that it disturbs submarine communication. Likewise, submarines have hidden in snapping shrimp colonies, which has made the animals unpopular with the US Navy.

Technological applications

Not only the snapping shrimp benefit from cavitating bubbles, but also the species *Homo sapiens*. Cavitation and collapsing bubbles play a crucial role in lithotripsy, the destruction of kidney or bladder stones with focused, strong ultrasonic pulses.

Probably the best known application of cavitating bubbles—at least for those who have their eyeglasses cleaned at the opticians—is ultrasound cleaning. For that application, a strong ultrasound horn is put into water. Bubbles cavitate in particular at surfaces, such as the eyeglass, and dirt particles are flushed away through microstreaming effects. Similar setups are used on a much larger scale for ultrasound cleaning in industry. Although a quantitative understanding of ultrasound cleaning has yet to be achieved, an ultrasound washing machine seems technologically possible.

Another important technological application of cavitating bubbles is sonochemistry, the enhancement of chemical reactions through ultrasound.[10] For some reactions, spectacular enhancement rates of several orders of magnitude have been achieved. The catalytic effect originates from the extreme temperature and pressure conditions inside the gas bubbles at collapse, which lead to dissociation of molecular gases. The resulting radicals trigger chemical reactions.

Light-emitting bubbles: Sonoluminescence

Cavitating bubbles, whether generated by ship propellers, in lithotripsy, in sonochemistry, or by snapping shrimp, disintegrate at bubble collapse because a shape instability develops. However, under other conditions, disintegration need not occur, and one can achieve controlled and stable cavitation. That phenomenon was discovered in 1988 by Felipe Gaitan, then a graduate student working with Lawrence Crum at the University of Mississippi. It became known as single-bubble sonoluminescence (SBSL; see the article by Crum in PHYSICS TODAY, September 1994, page 22).[8] In SBSL, a micron-sized bubble is acoustically trapped in a fluid-filled flask at resonance. Typically in water, the driving-pressure amplitude P_a is 1.2–1.4 bar, the driving frequency is 20–40 kHz, and the air saturation in the water is 20–40%. Once per cycle, at the Rayleigh collapse, the bubble emits a short pulse of light that typically lasts 100–300 ps. The origin of the light is thermal bremsstrahlung: At the adiabatic collapse, the gas inside the bubble gets heated, presumably up to about 15 000 K. Consequently, the gas partly ionizes, and, at recombination, light emission occurs.[8]

Although the energy at the bubble collapse is focused by about 12 orders of magnitude and the light emission is rather spectacular, the luminescence is negligible from an energy-balance point of view: The majority of the incoming acoustic energy is emitted again as sound (at the violent bubble collapse and, therefore, at much higher frequencies), converted into heat, or eaten up by chemical

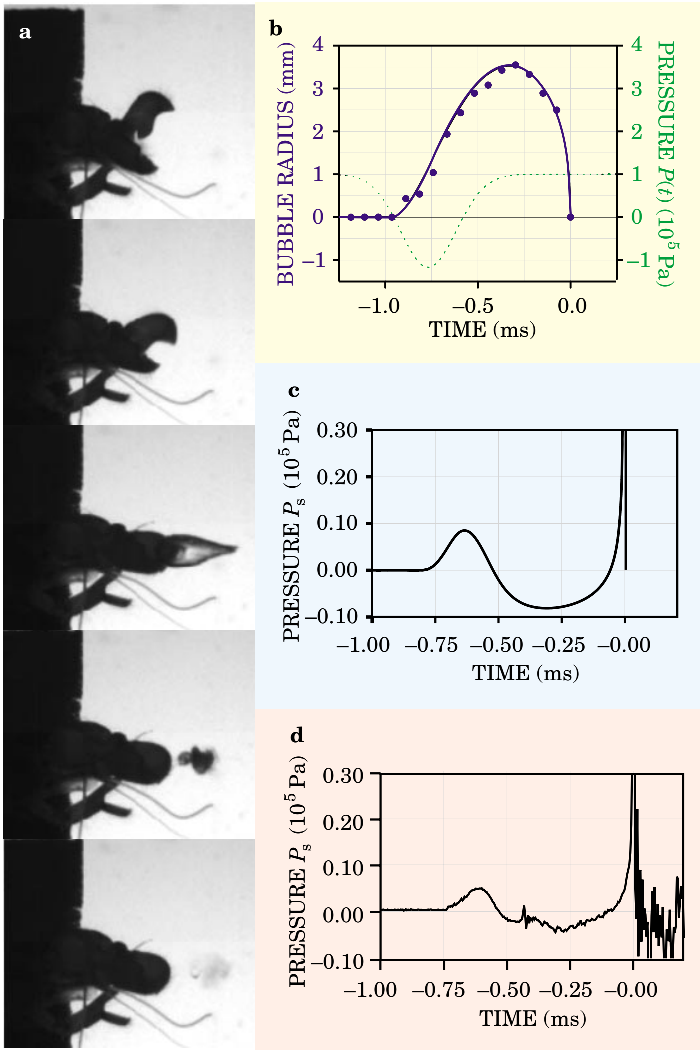


Figure 4. Snapping shrimp use bubble cavitation to stun and even kill prey. **(a)** Frames taken every 0.5 ms from a high-speed video recording capture the claw closure of a snapping shrimp. A fast water jet is emitted and generates cavitation. In the last frame, the bubble has collapsed; only microbubbles remain. **(b)** The measured and calculated bubble radius. The points are the measured data; the solid line shows the results from our Rayleigh–Plesset based theory (equation 4). The dashed line is the assumed pressure reduction $P(t)$ due to the jet. **(c)** The corresponding sound emission P_s from the bubble, calculated from equation 3. **(d)** The measured sound emission. (Adapted from ref. 9.)

reactions.[11,12] Therefore SBSL can be understood as "illuminated cavitating bubble dynamics," and indeed the discovery of SBSL gave cavitation physics a boost. The backbone of the theoretical understanding is again the Rayleigh–Plesset equation (equation 4).[7] Bubble dynamics as developed by Rayleigh, Milton Plesset, Andrea Prosperetti, and others determines the conditions under which stable SBSL can occur.[8] These conditions are (1) the collapse must be strong enough, that is, above the threshold for Rayleigh collapse to occur, (2) the bubble's shape must be spherical and stable, (3) the bubble must be diffusively stable, and (4) the bubble constituents must be chemically stable. Applying these criteria, the phase diagram of sonoluminescence can be quantitatively calculated, as shown in figure 5.

The chemical activity inside the bubble is a consequence of the high temperatures achieved. In fact, the sonoluminescing bubble can be viewed as a high-temperature, high-pressure microlaboratory or reaction chamber,[12] which can be controlled through external parameters such as forcing pressure, frequency, and dissolved gas concentration. When the bubble is expanding, liquid vapor and gas dissolved in the liquid are entering the bubble. At the adiabatic collapse, these gases are partly trapped inside the hot bubble, and they react. For example, nitrogen molecules will first dissociate to nitrogen radicals and later react to form NH, NO, and so on, which all dissolve readily in water when the bubble cools down and re-expands. Subsequently, the next reaction cycle starts.

Single-bubble sonoluminescence can be viewed as the "hydrogen atom" of cavitation physics. Single spherical-bubble cavitation is the simplest building block of a sound-driven bubbly fluid, just as hydrogen is for more complicated atoms, molecules, and solids. It is astounding how many subdisciplines of physics and chemistry have been necessary to understand the conceptually simple building block of a single bubble oscillating in a sound field: acoustics, fluid dynamics, plasma physics, thermodynamics, atomic physics, spectroscopy, chemistry, dynamical system theory, and applied mathematics in general.

Just as the understanding of heavier atoms, molecules, or solids poses more difficulties than that of hydrogen, so interacting bubbles, bubbles close to walls, and bubbly fluids are more difficult to understand than an isolated cavitating bubble. Figure 6 shows a picture of a col-

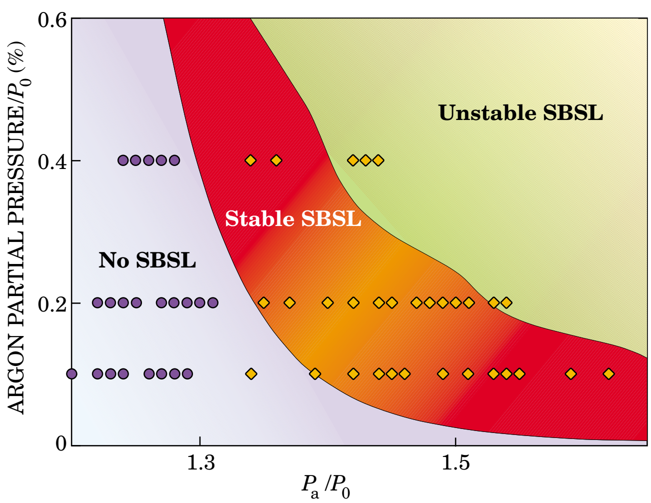


Figure 5. Phase diagram of single-bubble sonoluminescence (SBSL). The horizontal axis is the driving-pressure amplitude P_a, normalized by the ambient pressure P_0. The vertical axis is the argon partial pressure, also normalized by P_0. Stable SBSL is predicted to occur only in the red region. At lower forcing or Ar partial pressures, no SBSL occurs. Above the red region, the SBSL is unstable: The bubble grows by diffusion, and the phase and intensity of the emitted light aren't constant. The experimental data (dots in the respective color of the region) are in reasonable agreement with the predictions. (Adapted from ref. 8.)

lapsing bubble close to a wall. Due to the bubble's asymmetry, jets can develop that are directed toward the wall. Most of us have observed a related phenomenon: In champagne, bubbles tend to sit just under the fluid surface for some time. When the interface breaks, the cavity at the surface collapses and a jet forms, shooting little drops of champagne into the air.[13]

Figure 6. An asymmetrically collapsing bubble next to a wall. The bubble formed in water that was shaken at a frequency of about 60 Hz with the ambient air pressure near the liquid's vapor pressure. The maximal diameter of the bubble is about 1 mm. (Photo from L. A. Crum, *Journal de Physique Colloque* **40**, 285, 1979.)

Medical applications of bubbles

Understanding bubble–bubble and bubble–wall interactions is also crucial for many applications of bubbles in medicine. In the past few years, bubbles have become increasingly popular as contrast enhancers in ultrasound diagnostics.[14] In that technique, a solution of micron-sized bubbles is injected into the bloodstream. Normally the bubbles are coated to avoid clustering and to prevent surface tension from dissolving the bubbles by pushing the gas out of them. The bubbles scatter ultrasound (typically with a frequency around the bubbles' resonance frequency of 1–3 MHz) more efficiently than tissue or blood, and thus permit efficient flow visualization. For strong ultrasound, the bubbles also emit sound in higher harmonics. Those harmonics allow for better contrast to tissue, which scatters sound at mainly the fundamental frequency.

A very important application of bubbles in ultrasound diagnostics is reperfusion imaging of the myocardium, the heart muscle. Injected bubbles floating through the veins in the heart muscle scatter sound, which can be monitored. Applying a strong ultrasound pulse destroys the bubbles due to their shape instability. Correspondingly, the scattered sound signal nearly vanishes. After a second or so, however, new bubbles flow into the heart muscle, again giving a scattering signal. The time constant of the signal-recovery process yields information on potential heart damage.

A new trend in bubble medicine is to use the same kind of microbubbles for therapy, in which the bubbles can act as vectors for directed drug delivery and gene transfection into living cells. The permeability of cell walls for large molecules (both drugs and genes) is dramatically increased in the presence of ultrasound and microbubbles.[15] The nature of the mechanism behind this phenomenon is not yet understood. Jet formation, induced by collapsing bubbles, is one of the candidates for enhancing cell-wall permeation: Electron micrographs of insonated leukemia cells show conspicuous holes in their walls.[16] Jet cavitation damage and cell-wall permeation could thus be two manifestations of the same process. However, other high-energy processes besides jets are associated with the bubble collapse and could be important: Shear and pressure forces, sound waves, and shock waves also provide significant mechanical interactions between bubble and cell.

To further optimize the process of local drug delivery or gene transfection with the help of bubbles, it will be crucial to obtain a better understanding of both the hydrodynamic[2] and the acoustic forces acting on bubbles—in other words, to control the bubbles.[17] Given that even the dynamics of a rising bubble in still water is not fully understood, this task remains challenging.

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