

## Sonoluminescence from an isolated bubble on a solid surface

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Measurements of sonoluminescence from isolated cavities that spontaneously form on solid objects in a fluid-filled acoustic resonator are reported. The subnanosecond light flashes have a smooth ultraviolet spectrum similar to that from single bubbles, although with organic liquids low intensity Swan lines ride on the continuum. The hemispherical cavities reach maximum radii about five times larger than are realized in sonoluminescence from single bubbles. The sonoluminescence intensity continues to increase as the liquids are cooled to temperatures as low as 160 K. [S1063-651X(97)04112-3]

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We have studied a regime of sonoluminescence (SL) from an isolated cavity which spontaneously sets up as one increases the amplitude of sound in a resonator that contains a defect such as a wire or other solid body. Within experimental accuracy this cavity is a hemisphere (see Fig. 1) attached to the defect. The hemispherical bubbles emit a subnanosecond flash of light with each cycle of sound, and furthermore the spectrum is strongly weighted in the ultraviolet (see Fig. 2). The presence, in this complicated geometry, of these signatures of the extreme conditions expected to exist in SL from single spherical pulsating bubbles indicates the robust nature of the energy focusing mechanism at play in the fluid mechanics of bubbles [1]. The ease with which SL can be achieved in the arrangement reported here stands in contrast to the case of single spherical bubbles where the quality of resonator design plays an important role [1]. As the hemispherical bubbles are about ten times larger than the spherical bubbles, they are more amenable to experimental probes of the energy focusing and light-emitting mechanisms. In particular the hemispherical bubbles damage the boundary

on which they sit, and it may be possible to determine (with photography?) whether this effect is due to jets or shocks or some other mechanism.

Surface bubbles first appeared during our efforts to cool single xenon bubbles trapped in ethanol to its freezing point of about 160 K [2,3]. These experiments are a preliminary step toward obtaining SL in cryogenic systems such as would be realized for helium bubbles trapped in liquid argon. A variable-temperature optical cryostat was employed for these measurements, using liquid nitrogen as the coolant [4]. The measurements in Fig. 3 show that the increase in brightness with decreasing temperature for a 150-torr xenon bubble in ethanol seen in earlier studies [2] continues to hold even at considerably lower temperatures. As is known from research on SL, the increase in brightness is due to the temperature dependence of the upper threshold (the maximum drive amplitude allowed before the bubble disappears) [1]. The jump seen in the data near 220 K is accompanied by a switch from single bubble SL to a bubble on the surface of a piece of nichrome wire near the middle of the cell. Data for

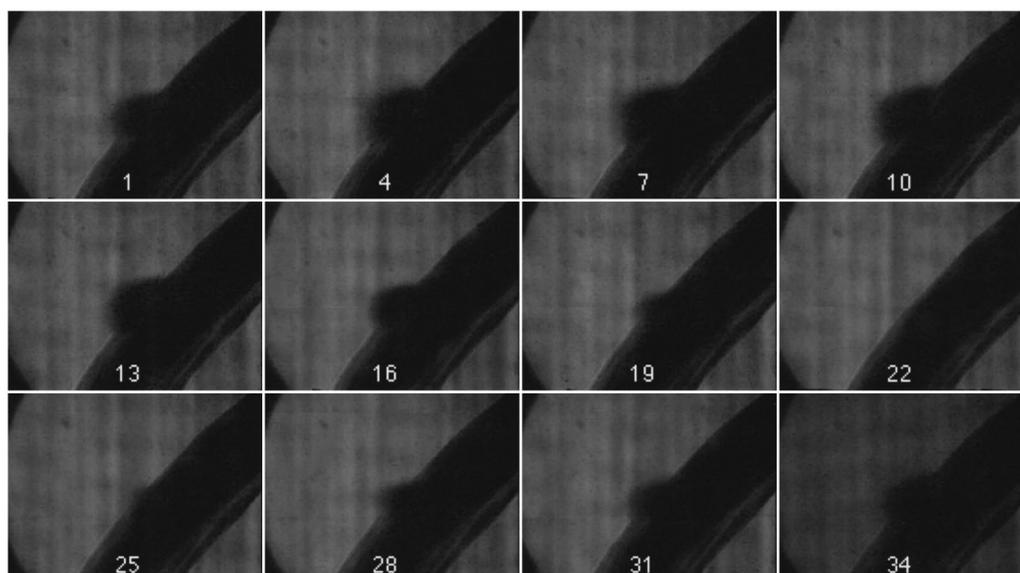


FIG. 1. Video images of one cycle of the oscillations of a 350-torr xenon bubble in water on a thermocouple wire. The numbers in the frames indicate the time in microseconds.

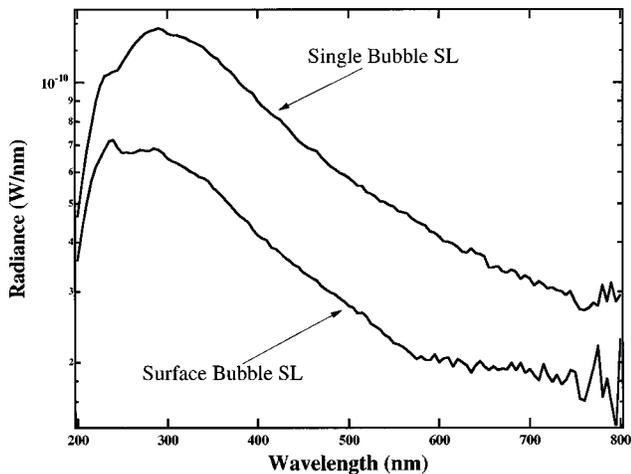


FIG. 2. Spectrum of 300-torr xenon in a water-surface-bubble and 150-torr 1% xenon in oxygen in a water-single-bubble SL generated in the same cell and acquired with the same equipment (10-nm full width at half maximum resolution). The dip at 240 nm and the falloff near 200 nm are attributed to absorption in the quartz wall of the resonator.

temperatures below 220 K were taken for these surface bubbles, but the maximum intensity was limited by the electronics and amplifier in the drive circuit. Subsequently, the experiments were greatly simplified by discovery of the same phenomena in silicone oil below  $-8^{\circ}\text{C}$ , and in *n*-dodecane and water at room temperature.

The remainder of the measurements described in this paper use xenon bubbles in water or *n*-dodecane driven by sound in a cylindrical quartz resonator sealed with stainless-steel endcaps. The endcaps were cooled so that the liquid in the system was maintained at  $12^{\circ}\text{C}$ . This cell is similar to those used for single bubble experiments with the modification that piezoelectronic transducers were mechanically com-

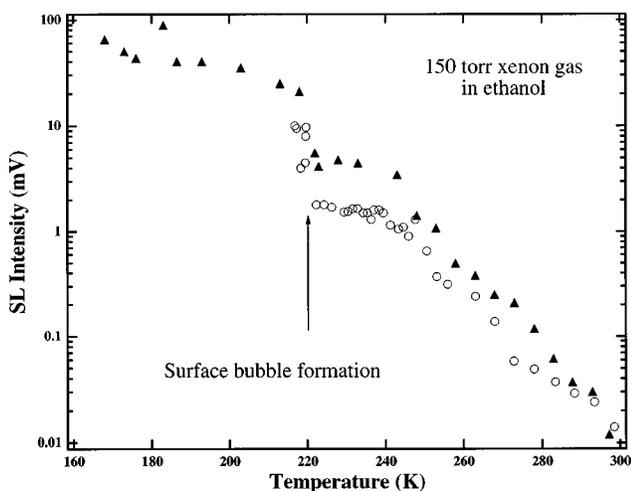


FIG. 3. Intensity of single-bubble SL as a function of temperature for 150-torr xenon in ethanol down to the freezing point of the alcohol. Below 220 K the data are for a surface bubble on a piece of nichrome wire. The maximum signal for a 150-torr air in water bubble in this system is about 6 mV. Data were acquired by collecting the SL light with a PMT whose output was detected with a lock-in amplifier referenced to the acoustic drive. The different symbols indicate runs on separate days.

pressed against the endcaps [5]. This allowed the sound amplitude in the cell (driven at 30 KHz) to be increased to levels where the hemispherical bubbles spontaneously form on a thermocouple inside the cell near the central pressure antinode.

The light from xenon bubbles is easily seen by the unaided eye in a darkened room, but a quantitative measure of the maximum intensity for this system is difficult to define. At some low threshold of drive pressure, a single bubble appears on a surface. As the sound intensity is raised, the light emission from this bubble increases until another threshold is reached, above which more bubbles appear on other surfaces, dramatically increasing the total light output of the cell. At still higher levels, brief cavitation events which emit faint pulses of light are seen in the bulk of the liquid. The maximum brightness from the single surface bubble of 300-torr xenon in the water level is about 1–2 times 150-torr air in water single-bubble SL at room temperature. The heavy noble gases seem essential for this type of SL. Emission at levels about half of xenon have been seen with krypton gas dissolved in the water. With argon in water, the surface bubbles produce barely detectable signals. Although the spontaneous appearance of bubbles on the wire at high sound amplitude occurred in a very similar fashion when sulfur hexafluoride, oxygen, helium, hydrogen and air were dissolved into the water, no detectable light emission was present. By using lower drive amplitudes, single-bubble SL could be seeded in the same cell by pulsing a current through a nichrome wire inside the cell. For helium, argon, and air, typical single-bubble SL was observed. In summary, the isolated surface bubbles occur at drive levels higher than those which maintain single spherical bubbles, yet lower than the level at which full blown cavitation appears.

Images of the hemispherical pulsations shown in Fig. 1 were acquired with a video technique similar to that demonstrated by Tian, Ketterling, and Apfel, and used by Holt and Gaitan [6]. The bubble was backlit with a light-emitting diode (LED) that produced a 500-ns flash of red light that was driven by a delay generator triggered on the synchronous output of the sine wave generator used to excite sound (at 30 KHz) in the resonator. A  $60\times$  microscope (Edmund Scientific) focused the shadow onto a video camera. The electric iris in the camera was set so that each video frame was an integration of  $\frac{1}{250}$  s. Therefore each video frame was an integration of about 120 sound cycles all at the same phase. By stepping the phase of the delay generator driving the LED, the full cycle of motion was imaged. The radius time curve in Fig. 4 is taken from this data, and calibrated by the magnification of an object of known size.

As discussed by Jones and Edwards, and noted by Benjamin and Ellis [7], the Rayleigh-Plesset (RP) equation [8] is spherically symmetric, and hence applies to the case of a hemispherical bubble on a semi-infinite solid boundary, provided that the viscous effects at the interface are minor. In the spirit of this model we have compared the data in Fig. 4 to solutions of the RP equation. The failure of the imaging optics to resolve the smallest bubble sizes limits the accuracy of the fit but reasonable agreement is found for expansion ratios ( $R_{\text{max}}/R_0$ ) of 10:1 or 20:1 and drive pressures  $P_a$  of 3–5 atm. A direct measurement of the drive pressure was difficult due to the sealed geometry of the resonator, and the

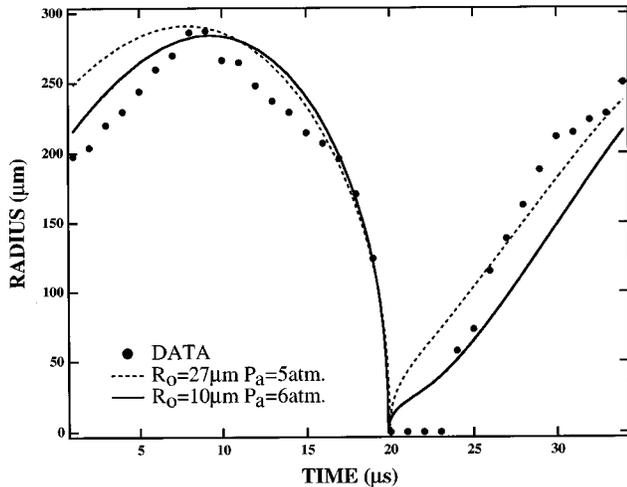


FIG. 4. Radius vs time data for the bubble in Fig. 1, with solutions of the Rayleigh-Plesset equation for comparison. The sound period is  $33.6 \mu\text{s}$ . For the frames at  $20\text{--}23 \mu\text{s}$ , no image of the bubble could be resolved in the data of Fig. 1.

tendency for cavitation to occur on the hydrophone. By inserting a hydrophone down the fill tubes, sound amplitudes of 3.5 or 4 atm have been measured under conditions producing the wire bubbles. These fits to the RP equation imply a significant scaling up in the size of sonoluminescing bubbles. These surface bubbles oscillate between a maximum radius of about  $250 \mu\text{m}$  and a minimum radius (implied by the RP simulations) on the order of a few  $\mu\text{m}$  (instead of the sub- $\mu\text{m}$  values typical of single-bubble SL). The hemispherical bubbles may be well described by the Rayleigh-Plesset equation because the viscous penetration depth of  $3\text{--}5 \mu\text{m}$  is small compared to ambient and maximum radii.

Additional evidence for Rayleigh-Plesset-type dynamics is found in light scattering data. To obtain the data in Fig. 5, light scattered from a 1-mW HeNe laser beam trained on a wire bubble is collected from around  $60^\circ$  from the forward direction by a short focal length lens, and focused through an aperture onto a photomultiplier tube (PMT) (Hamamatsu R2027) [9]. The output of the PMT was acquired in real time by a digital oscilloscope. Because of the large level of dc scattering from the wire, as well as the lack of a good deconvolution function from the light amplitude to a radius, actual radius time data could not be extracted from these curves, but the general relationship of a more negative signal, indicating a larger radius, should still be valid. The bifurcations and period doubling seen in the data in Fig. 4 are well known features of the RP equation for large bubbles or high levels of drive [10].

Using the light scattering technique, the timing of the flash with respect to the bubble dynamics (350-torr xenon in water) was measured in two stages: (i) the laser scattering was acquired with the sound sync as the trigger; and (ii) with the laser off, the sync was acquired by triggering acquisition on the SL pulse, as detected from the bubble by a lens focused through an aperture onto a PMT. The lensing insured that the SL was emitted by the same bubble that the laser illuminated. By aligning the sound sync in these two pieces of data, the relative phase of the bubble motion and the SL

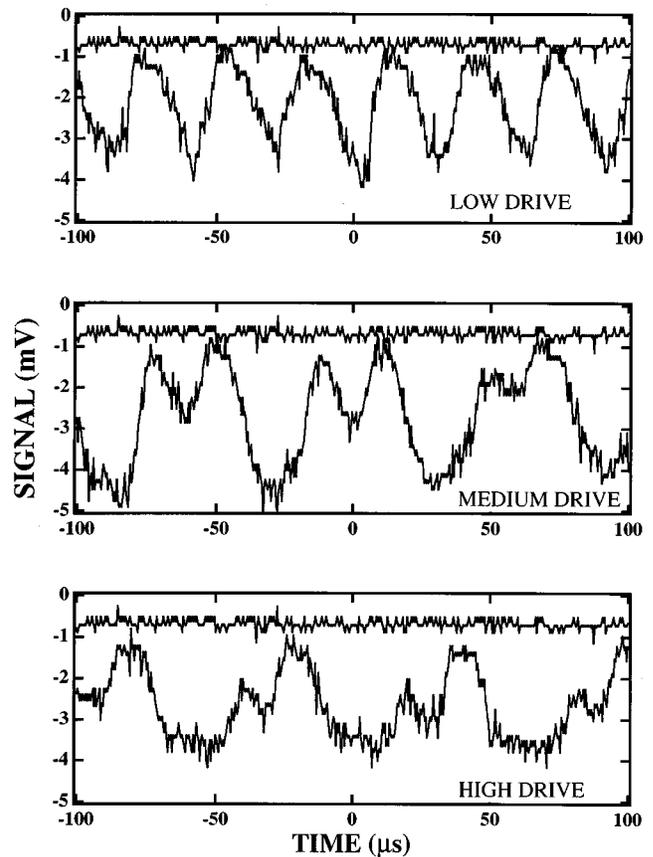


FIG. 5. Laser scattering measurements of the oscillations of a 350-torr xenon in a water-surface-bubble as a function of drive pressure. The data are single-shot measurements of the PMT signal due to scattering from the bubble. A noise floor acquired with no bubble present is also shown. Note the bifurcations and period doubling in the dynamics. In the bifurcated dynamical states it was observed that the larger collapses were accompanied by a flash of light, while the softer collapses generated no light.

flash could be deduced. The flash generally occurs near the minimum radius but due to jitter in the system the exact phase was uncertain over the range of  $5\text{--}10 \mu\text{s}$ . This uncertainty illustrates how the flash to flash synchronicity of this system is reduced from that of single-bubble SL. This may be due to bubble-bubble interactions, bubble-surface interactions, or parasitic modes of the cell transiently excited by the high level of drive present.

Although the jitter in the time interval between flashes is greater than for spherical bubbles, these spontaneously generated bubbles still produce flashes with subnanosecond time scales similar perhaps to single bubble SL [11]. The response of a 650-ps rise time PMT (Hamamatsu R5600U-03) to flashes from single-bubble SL, and from the wire bubble (350-torr xenon in water), are both instrument limited. We conclude that the flashes from the wire bubble are comparable to or shorter than the impulse response of the detector [12].

The spectrum of the light from the surface bubbles is also very similar to that of single-bubble SL. Using techniques described elsewhere [13], the data shown in Fig. 2 were measured for a 300-torr xenon in water bubble on the thermocouple surface, as well as a single-bubble spectrum from the

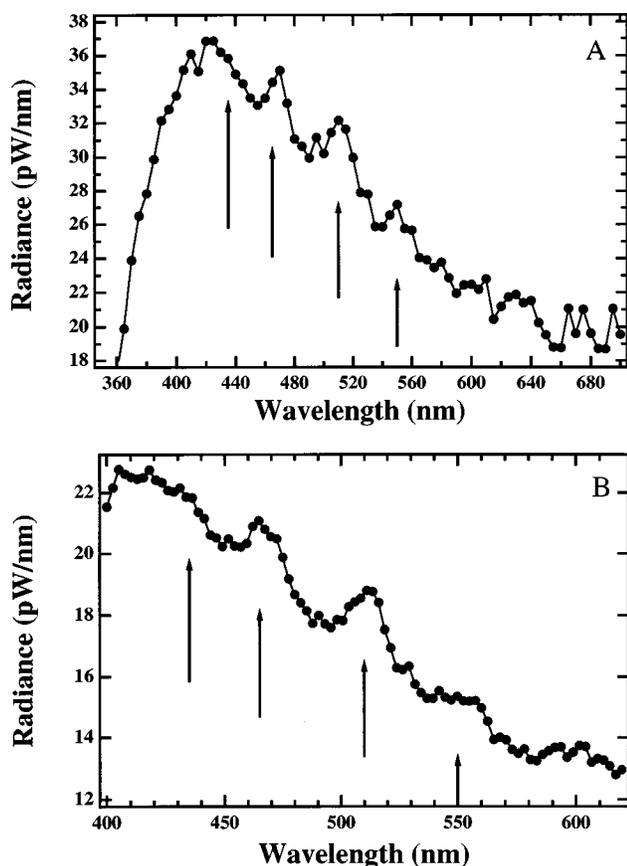


FIG. 6. Spectrum of 290-torr xenon in *n*-dodecane with 10-nm full width at half maximum resolution. The cutoff in blue is consistent with absorption in the liquid. Shown in (B) is a second scan of the Swan lines with the same resolution. The arrows indicate the expected location of Swan lines.

same resonator for comparison. It is generally broadband increasing into the UV right up to the extinction cutoff due to the water. The upturn in the red is a notable unexplained difference from single-bubble SL, but was consistently seen for the surface bubbles. The long-wavelength enhancement could be due to preferential reflection from the surface for red light initially emitted in the opposite direction from the spectrometer, or one could imagine it as a few thousand degree black body superimposed on the usual SL spectrum. In fact, these bubbles occur on a thermocouple, and it was noticed that the temperature recorded for the junction which was millimeters away from the bubble would rise  $10^\circ$  when the bubble was present, and as soon as it was gone would return to the ambient liquid temperature in the silicone oil system (which has a low thermal conductivity). In an attempt to see a faint 1000–2000-K blackbody spectrum, data were taken of a non-light-emitting oxygen bubble but no signal in the red (or anywhere between 200 and 800 nm) was seen.

The existence of a hint of spectral lines due to excited states of molecular carbon (Swan lines) in the xenon in dodecane system (see Fig. 6) points toward some connection with these surface bubble and transient multibubble SL [14]. These spectral lines are much less prominent than in multibubble studies, and in fact are barely visible over the continuum spectrum that is characteristic of single-bubble SL. In an effort to look for other spectral lines, the wire bubble was

run in salt water without any evidence of the sodium or  $\text{OH}^*$  lines seen in multibubble studies [15]. To investigate excimer transitions as an explanation of why xenon and krypton are the only gasses to give light for the surface bubble systems, we sought to induce a known excimer transition between xenon and chlorine (whose energy is 308 nm) by running the wire bubble with xenon gas dissolved into a mixture of water and sodium hypochlorite (household bleach). This liquid has a strong absorption in the spectral region of interest, but there was signal detected smoothly through the absorption, and no evidence of excimer activity or anything other than the usual smooth single-bubble sonoluminescence spectra was seen.

Although all results reported here were measured for bubbles on the surface of a piece of wire, the same phenomena has been seen on quartz, sapphire, stainless steel, and copper foil. Bubbles on the sapphire window open up the possibility that, if there is a sufficiently small amount of liquid between the bubble and the window, then by using vacuum spectroscopy (to eliminate absorption due to the atmosphere) perhaps the spectral nature of the emission could be investigated below the 185-nm cutoff imposed by the water (which is the most UV friendly fluid studied to date). Cavitation damage [16] has been seen in conjunction with these hemispherical bubbles as well. For example, a  $2 \times 2\text{-cm}^2$  piece of 0.002-in.-thick copper was suspended near the pressure antinode at the center of the cell. With high levels of sound, light-emitting bubbles were seen all over its surface. After about 1 h of continuous running the surface was scarred and a  $1 \times 1\text{-cm}^2$  hole was cut into the foil with small fragments of copper present on the bottom of the cell.

The cavitation damage as well as the collapse of the bubble close to a boundary suggests a possible role for jets in SL, as has been previously suggested by Prosperetti and Longuet-Higgins [17]; however, the existence of a jet is not absolutely guaranteed by these observations. Experimental and numerical investigations of jets usually involve bubbles which begin to collapse with their center at least one radius from the boundary (although there are few studies of bubbles with parameters similar to the ones seen here) [18]. Jones and Edwards [5] generated hemispherical caps on a surface much like the images in Fig. 1, and did not report evidence of jets. The application of Rayleigh-Plesset theory to the hemispherical bubbles keeps alive the potential for (hemispherical?) shock waves inside these bubbles [19]. It is hoped that future optical microscopy experiments with these scaled up bubbles on a transparent boundary of a resonator might allow direct imaging of whatever process is occurring in the light-emitting region in the bubble. Such measurements do not seem feasible for single-bubble SL, since the radius of the bubble at the light-emitting moment is under  $1 \mu\text{m}$  [1,9,20].

It is remarkable that in a simple system such as spontaneous cavitation of a liquid, nature chooses the runaway solution of hydrodynamics that provides picosecond flashes of UV light just as in the more controllable phenomena of single-bubble SL. Even in complex geometries one can marvel at SL by simply introducing a solid object into the sound field and turning up the drive. The failure of gases other than xenon or krypton to emit light remains unexplained, and probably accounts for the lack of light production from surface bubbles in earlier cavitation experiments but is consistent with experiments on flow induced cavitation [21].

Although the great synchronicity of single-bubble SL is diminished for the surface bubbles, the presence of the boundary and the scaled-up size of these bubbles as well as the easy method of generation may lead to experiments which will uncover the mechanism responsible for the emission of light.

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