

Physical aspects of sonoluminescence from acoustic cavitation

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The passage of an ultrasonic wave through a medium can sometimes result in the production of acoustic cavitation which is often accompanied by the generation of light; a process known as sonoluminescence or 'light from sound'. A brief historical overview of early sonoluminescence (SL) research is presented, followed by a phenomenological description of the physics of cavitation bubble fields. Many of the published results describing the features of SL (pulse duration, phasing, spectra, etc.) are inconsistent and sometimes contradictory. However, recent studies of sonoluminescence from individual, radially pulsating bubbles draw a consistent picture. Single-bubble SL flashes occur once *every* acoustic cycle and coincide precisely with bubble collapse. The duration of the flash is believed to be ≤ 50 ps and the emission spectra indicate substantial heating of the bubble interior. These results suggest that the primary mechanism for single-bubble sonoluminescence is the rapid heating of the interior of the collapsing bubble.

Keywords: sonoluminescence; cavitation; single-bubble flashes

When an acoustic wave propagates through a liquid containing microscopic gas inclusions, these 'nucleation sites' can be mechanically activated, at which point they spawn free bubbles which then undergo highly energetic volume pulsations. Associated with these pulsations is a broad range of linear and nonlinear mechanical behaviour, the nature of which will depend primarily on the acoustic pressure amplitude and the equilibrium (i.e. undisturbed) bubble size relative to the acoustic wavelength. This activity, which is loosely termed acoustic cavitation, is often accompanied by electromagnetic emissions consisting of energy in the UV and visible bands. The physical basis for this process, which is termed *sonoluminescence*, is the topic of this paper.

One of the remarkable features of acoustic cavitation lies in its ability to concentrate mechanical energy. For example, it is well known that sonoluminescence (SL) can be generated in water by acoustic waves with peak amplitudes of order 1 atmosphere. Such an acoustic pressure corresponds to an energy density of 2.2 J m^{-3} or, in a less conventional unit, approximately $4 \times 10^{-10} \text{ eV molecule}^{-1}$. In contrast, there is recent evidence that photons associated with sonoluminescence possess energies in excess of 6 eV; thus, the generation of SL from an acoustic wave implies an energy density 'amplification' (or concentration) of roughly 10 orders of magnitude! This is comparable to the energy gain observed when a thermal neutron is absorbed by the fissionable isotope of uranium.

Sonoluminescence was first observed nearly 60 years ago^{1,2}, and since then there have been a variety of

explanations given for the electromagnetic emissions. Electrical microdischarge theories of various types have been quite popular. In the balloelectric model, Harvey³ proposed that an electric field present in the liquid gains strength, and ultimately discharges, as the bubble collapses. In 1940, Frenkel⁴ suggested that electrical charges known to exist on the surface of the bubbles were somehow made to discharge, probably during the expansion phase of the bubble pulsation. This theory has been challenged by the experiments of Meyer and Kuttruff⁵, Kuttruff⁶ and Suslick^{7,8}. In their 'hot-spot' model, Noltingk and Neppiras⁹ suggested that SL stems from the adiabatic heating of the gas in the collapsing bubble. In a sense, the gas is heated to incandescence. More recently, a new electrical microdischarge theory proposed by Margulis has shown some promise^{10,11}. Although the results summarized below tend to support the thermal theory (or variations thereof), it is clear that the exact origin of these emissions is not fully understood.

Much of the published work in sonoluminescence has involved attempts to obtain the emission spectra and through this learn something about the cavitation process itself. In their original treatise, Noltingk and Neppiras⁹ proposed that luminescence results from black-body radiation of hot gas. However, compressional heating can also lead to molecular emission associated with chemical species in the gas and/or vapour. Taylor and Jarman¹² discovered that an aqueous solution of NaCl yielded a SL spectrum which possessed peaks corresponding to the sodium D-lines. From this they inferred the bubble collapse temperature to be in the order of 10^4 K . Sehgal

*et al.*¹³ also examined the process in a variety of liquids and obtained values ranging from 3040 to 3705 K. This approach has been greatly refined by Suslick and his associates^{7,8,14-16}, who have made significant contributions to sonochemistry through their study of SL emissions in cavitation fields. Through careful experimental design, they can identify individual transitions that contribute to specific peaks in the spectrum and thereby fit the spectrum using a single adjustable parameter – the ‘temperature’ of the cavitation collapse. Using this approach, they predict sonoluminescence temperatures in the order of 5000 K¹⁷.

Sonoluminescence from a cavitating bubble field

Many, if not all, of the aforementioned experimental efforts involved the study of light emanating from a cavitation-bubble field. For example, Suslick *et al.* generated cavitation using a high power acoustic horn immersed in a liquid. In such a system, one observes copious cavitation at the tip of the horn and, in some cases, throughout the bulk of the fluid¹⁸. Cavitation fields exhibit extremely complex dynamical behaviour. Gas and/or vapour bubbles of various equilibrium sizes pulsate at various phases relative to the driving acoustic pressure field, often interacting with each other both acoustically and hydrodynamically. On a microscopic level, the cavitation is seen to be transient in the traditional sense, appearing and disappearing spontaneously. Macroscopically, however, there often appears to be a cyclic nature to the cavitation process in which bubbles periodically agglomerate in regions of dense cavitation activity just prior to being ejected by the same acoustic radiation-pressure forces that drew them together in the first place. Not surprisingly, these regions of intense cavitation are also sites of maximal luminosity¹⁸.

It is well-known that the driven, radial pulsations of a cavitation bubble can result in significant heating and compression at the point of collapse^{9,19,20}. However, bubbles in cavitation fields rarely pulsate radially. The presence of nearby bubbles and/or boundaries perturb the otherwise radially symmetrical flow field, resulting in an asymmetric collapse and the subsequent formation of a liquid jet that penetrates the centre of the bubble^{21,22}. Collapse asymmetry tends to reduce the maximum bubble temperature while the formation of a jet introduces liquid into the interior of the hot bubble. In addition, most experiments involving sonochemistry in cavitation fields utilize liquids that are either saturated with gas or only slightly degassed. Gas in a cavitation bubble ‘cushions’ the collapse, thereby lowering the collapse pressure and temperature. Moreover, at low to moderate acoustic pressure amplitudes, the dynamical response of a gas-filled bubble tends to be stiffness dominated. That means that there is a sensitive relationship between the phase of the bubble motion and the equilibrium bubble size relative to the acoustic wavelength. Since cavitation fields possess bubbles of varying equilibrium sizes, one can expect a range of dynamical responses.

The plethora of complex bubble behaviour described above generally results in a faint sonoluminescence glow which is sometimes visible to the naked eye. Depending on experimental conditions, individual light flashes can be poorly correlated in time. Measured lifetimes for

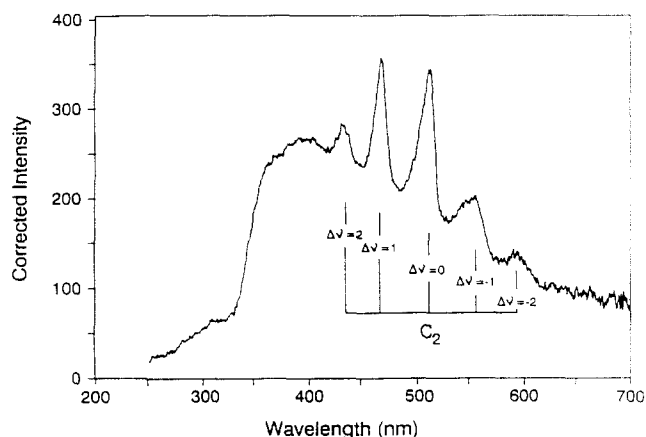


Figure 1 Sonoluminescence from cavitated dodecane under argon at 4 °C. The spectral peaks correspond to the various rotational and vibrational bands of diatomic carbon (from Suslick *et al.*¹⁴)

SL flashes range from nanoseconds^{10,12,23} to microseconds^{24,25}. Reports of the phasing of the SL flash vary widely as well, with published observations of light emission during the compression phase⁵ as well as the expansion phase¹¹. In general, reductions in either the thermal conductivity of the gas^{26,27} or the vapour pressure of the liquid²⁸ result in enhanced luminosity – observations which support a thermal model for sonoluminescence. Furthermore, the measured SL spectra are typically characteristic of the liquid rather than the gas. For example, *Figure 1* shows the SL spectrum, obtained by Suslick *et al.*, from a cavitation field in dodecane under argon at 4 °C¹⁴. The peaks correspond to various rotational and vibrational bands in diatomic carbon. Suslick *et al.* have refined the interpretation of sonoluminescence spectra to the point where they can predict bubble collapse temperatures with precision¹⁷.

Synchronous, single-bubble sonoluminescence

The theoretical modelling of bubble motion in a cavitation field is complicated by the fact that most theories describing nonlinear bubble dynamics assume purely radial oscillations. Until recently, there has been little or no experimental evidence of light emanating from a single, radially pulsating bubble. In 1988, while studying the conditions necessary for sonoluminescence in a cavitation field, Gaitan *et al.* discovered that, for degassed liquids, a single, stably pulsating bubble could be made to produce a SL flash each and every acoustic cycle²⁹. They utilized a technique known as acoustic bubble levitation, a process in which the radiation stress on the surface of a bubble in an acoustic standing-wave field offsets the gravitational buoyancy and serves to levitate the bubble at a fixed position far removed from solid boundaries³⁰. The standing wave controls the position of the bubble and drives it into radial pulsation.

Simplified block diagrams of the experimental arrangements are shown in *Figure 2*. The test liquid was a degassed and filtered mixture of distilled water and glycerin (21% by weight). By measuring the scattering of laser light off an acoustically levitated bubble, they were able to determine the bubble radius *versus* time curve in relation to the acoustic pressure; see *Figure 2a*. In a separate measurement involving the same bubble, they

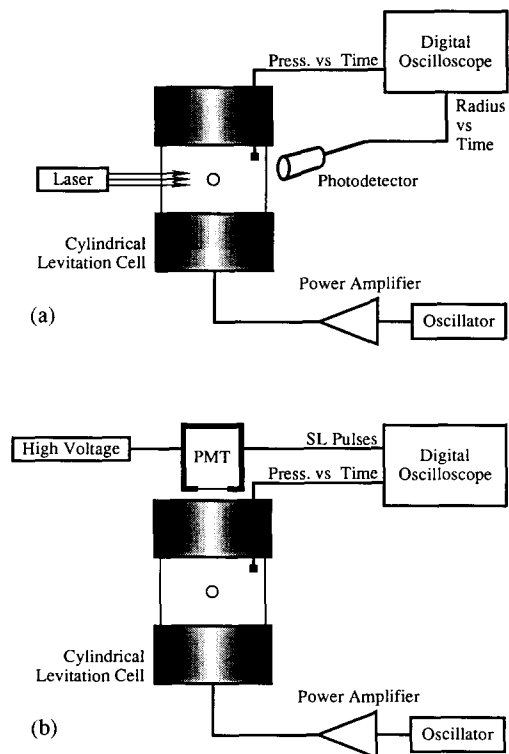


Figure 2 Simplified block diagrams of the experimental apparatus employed by Gaitan *et al.*²⁹. The cylindrical resonator was driven in the $r, \theta, z = 1, 0, 1$ mode at a frequency of ≈ 22 kHz. (a) Scattering experiment utilizing a linearly polarized, 3-W Ar-ion laser (wavelength = 488 nm) and a photovoltaic silicon photodiode detector. (b) SL measurements employing a Hamamatsu R585/C617 PMT/preamplifier unit designed for single photon counting. The digital oscilloscope operated at $100 \text{ M samples s}^{-1}$ with a conversion gain of 8 bits

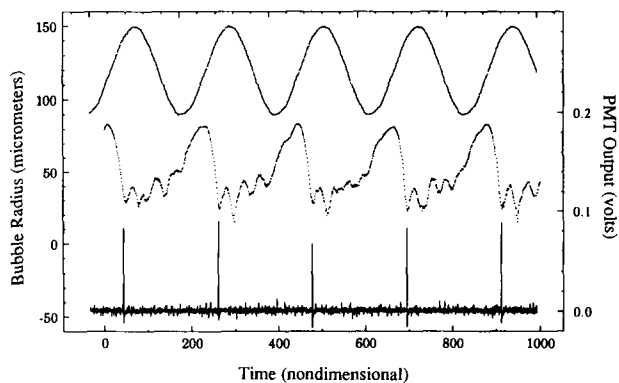


Figure 3 Synchronous relationship between the driving acoustic pressure (top trace), the bubble radius *versus* time curve (middle trace) and the sonoluminescence flashes (bottom trace) for a gas bubble with an equilibrium radius of $\approx 25 \mu\text{m}$ driven at an acoustic pressure amplitude of 0.12 MPa and at a frequency of 22.3 kHz. The liquid was a degassed solution of glycerin and water (adapted from Gaitan *et al.*)

utilized a photomultiplier tube (PMT) to measure the timing of the SL flashes in relation to the acoustic pressure; see Figure 2b. By using the acoustic pressure as a reference, they were able to temporally relate SL flashes to the bubble radius *versus* time. Figure 3 is a plot of the measured bubble radius *versus* time along with the accompanying sonoluminescence flashes and the acoustic pressure driving the bubble²⁹. It is evident that

the flashes are always generated while the bubble is at or near its minimum size. This is incontrovertible evidence in support of a thermal origin for SL generation.

This work was then followed by a set of remarkable measurements by Putterman *et al.* at UCLA which focused on the characteristics of the sonoluminescence flash itself^{31–33}. They utilized an acoustic standing wave to localize and drive a single bubble into stable radial pulsations in degassed distilled water. SL was monitored using the fastest available microchannel plate photomultiplier tube (Hamamatsu R2809U). Figure 4 shows the normalized PMT output current which resulted from a single SL flash³². Also plotted in the normalized PMT output when illuminated by a 34-ps pulsed laser operating at 410 nm, which matches the blue colour of SL. Note that the signals are virtually indistinguishable; it is impossible to tell which pulse is shorter. (The shape of the measured pulse is dominated by the response time of the PMT itself.) This led Putterman to conclude that the light pulse associated with single-

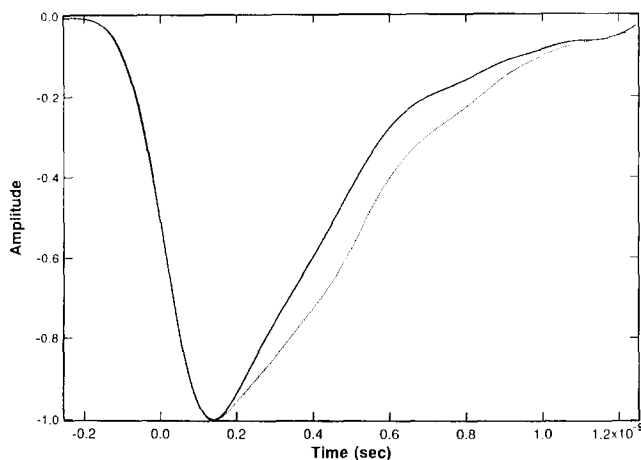


Figure 4 The current output from the microchannel plate PMT when excited by a sonoluminescence flash (solid line) and the output from a 34 ps pulsed laser (dotted line). The SL source was a radially pulsating air bubble in degassed water driven at 29.8 kHz. The tail in the response of the PLP compared to the SL flash is thought to be due to ringing in the laser cavity (from Barber *et al.*³²)

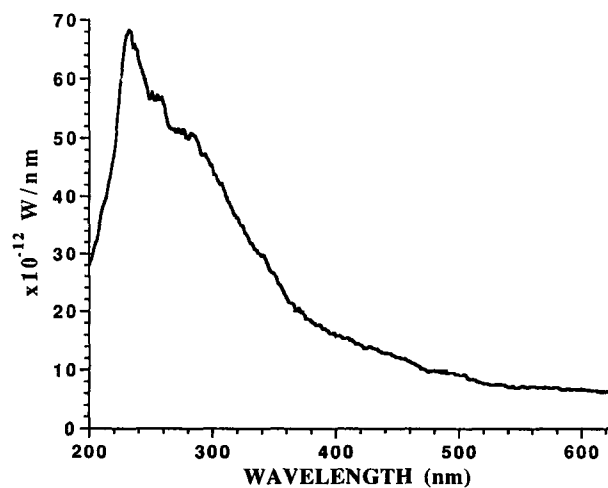


Figure 5 Calibrated spectral density of the synchronous SL flashes from a radially pulsating air bubble in degassed water at 22 °C. The acoustic driving frequency was 27 kHz. Note the wavelength scales in a comparison with Figure 1 (from Hillier *et al.*³³)

bubble sonoluminescence is no longer than ≈ 50 ps. It is important to point out that the pulse profile shown in Figure 4 is extremely repeatable and synchronous relative to the driving pressure field. In fact, repeated measurements of the time interval between successive DL pulses are normally distributed about a mean value of $33.5538 \mu\text{s}$ (the acoustic driving frequency was ≈ 29.8 kHz) with a standard deviation of only 50 ps³². As for the phasing of the flashes, laser scattering and SL measurements similar to those of Gaitan *et al.*²⁹ indicate that light is emitted ≈ 20 ns prior to the instant of maximum bubble collapse³⁴ which once again clearly supports the thermal theories.

As a follow-up to their light-pulse measurements, Hiller *et al.*³³ went on to measure the spectrum of single-bubble sonoluminescence. The calibrated spectral density of a sonoluminescing air bubble in water at 22 °C is given in Figure 5. The fall-off in the spectrum is claimed to be the result of UV absorption in the experimental apparatus (water, container, etc.) Calibration was achieved by comparison with a deuterium light source. Hiller *et al.* found that this measured spectrum deviated by only 10–20% from the tail of an 18 000 K black-body radiator. Similar spectra obtained at 10 °C show good agreement with a 30 000 K black-body spectrum! It appears that reducing the liquid temperature lowers the vapour pressure which in turn leads to a more energetic bubble collapse. It is important to note that these results are based on a rather extreme extrapolation of the data to undetectably short wavelengths. (There are those who feel that the spectrum of Figure 5 is most likely the result of pressure-broadened molecular emission associated with the excited water molecule and the hydroxyl radical³⁵.) It is evident that more data is required, possibly using liquids that readily admit UV transmission.

Regardless of the specific origin of the spectrum given in Figure 5 this spectrum is consistent with the notion of a hot bubble, in agreement with Noltingk and Neppiras¹¹. It remains to be seen, however, if the adiabatic heating of gas in a collapsing bubble can adequately explain the very short duration light pulses associated with synchronous, single-bubble sonoluminescence. When the velocity of the collapsing cavity wall exceeds the velocity of sound in the gas, a converging shock front is formed. Jarman²³ was the first to suggest that such shock waves may play an important role in SL production. Drawing from their study of the threshold acoustic pressure required for the onset of sonoluminescence in different gases³⁶, Vaughan and Leeman argued that shock waves may be more important than adiabatic compression in the early stages of bubble heating³⁷. Indeed, the possibility of shock-induced picosecond SL has drawn renewed interest in the wake of the recent discovery of single-bubble sonoluminescence.

Conclusions

Many of published results describing the features of SL (pulse duration, phasing, spectra, etc.) are inconsistent and sometimes contradictory. These studies were, for the most part, carried out in cavitation fields in which the bubble behaviour is very complex and difficult to

characterize. (Despite this fact, Suslick *et al.* have had good success in modelling the molecular transitions associated with the collapse of a hot bubble.) On the other hand, recent studies of sonoluminescence from individual, radially pulsating bubbles draw a consistent picture. Single-bubble SL flashes occur once every acoustic cycle and coincide precisely with bubble collapse. The duration of the flash is ≤ 50 ps and the emission spectra indicate substantial heating of the bubble interior. These results suggest that the primary mechanism for single-bubble sonoluminescence is the rapid heating of the interior of the collapsing bubble, either through adiabatic compression or the formation of a converging shock wave.

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