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Sonoluminescence from the unstable collapse of a conical bubble

T.G. Leighton [a],*, W.L. Ho [a], R. Flaxman [b]

[a] *Institute of Sound and Vibration Research, University of Southampton, Highfield, Southampton, SO17 1BJ, UK*
[b] *PCS Workshop, Cavendish Laboratory, Madingley Road, Cambridge, CB3 OHE, UK*

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Abstract

An experimental bubble collapse is designed such that a gas bubble expands slowly into a conical hollow through a reduction in the static pressure. When this pressure is rapidly released, the resulting liquid shock causes the bubble to collapse into the cone. Ideally a radial divergence or convergence of the liquid in the cone will follow the same geometry as that for a spherical pulsating bubble. Pressure transducers within the liquid at the base of the cone and at the cone apex, are able to detect rebound shocks and the pressures at the centre of the bubble, respectively. Sonoluminescence is detected from the collapse. © 1997 Elsevier Science B.V.

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1. Introduction

Inertial collapse of a spherical gas bubble in a liquid is characterized by a relatively slow, approximately isothermal growth of an initial bubble nucleus to many times its original size. This is followed by a rapid collapse, the initial stages of which are dominated by the inertia of the spherically converging liquid. During the collapse, the gas temperature rises as it is compressed, and shocks can propagate within the gas. Both high temperatures [1] and shocks [2] can potentially generate free radicals, the subsequent radiative recombination of which can emit light, termed sonoluminescence. There are, however, a range of proposed mechanisms [3–9] by which sonoluminescence can be generated. The production of this emission is sometimes taken as indicative of (but not necessarily a requisite for) the occurrence of inertial cavitation.

After reaching minimum size, the bubble rebounds, emitting a pressure pulse into a liquid. Until recently, the bubble was then thought to fragment. However, in 1990 Gaitan and Crum [10] discovered sonoluminescence over measurement intervals of thousands of acoustic cycles from repeated collapses of a single bubble which apparently did not break up. The discovery that the sonoluminescent flash in such circumstances is of less than 50 ps duration [11] throws into question the mechanism by which luminescence is generated in such circumstances. However, in practical applications involving the cavitation of many bubbles, the breakdown of spatial symmetry would suggest that each bubble performs such inertial collapses once or only a few times and then fragments on rebound. It may be that one or other of the various mechanisms for generating light from bubble collapse dominates, depending on the details of the collapse.

In this paper, the collapse of a single bubble is reported, and sonoluminescence is observed. However, the collapse is specifically designed to be unstable, so that the bubble can be ensured to have undergone fragmentation after rebound. In addition, the geometry of the bubble is such that it collapses into the solid angle conical section (30° half-angle) of a sphere. This not only allows the imaging of a ‘cross-section’ of the luminescing bubble, but also allows the positioning of pressure transducers within the gas and within the surrounding liquid, since the centre of the collapse is well defined.

2. Materials and methods

The basic apparatus consists of a steel U-tube, of 60 mm internal diameter, partially filled with degassed

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* Corresponding author. E-mail: tgl@isvr.soton.ac.uk; fax: +44-1703-593190.

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water (Fig. 1). The internal pressure within the tube can be reduced through connections to a vacuum pump above the level of the liquid in the longer leg of the tube, which is terminated by a spring-loaded top plate. Once the valve to the vacuum pump is closed, the raising of this plate using the lever returns the pressure exerted on the tube contents to atmospheric. The shorter leg is terminated by a transparent hollow cone of 30° half-angle. When under atmospheric pressure, this leg and cone are filled with water, except for a bubble of millimeter-order diameter which is injected into the apex of the cone down a length of rubber tubing which is temporarily fed through the length of the U-tube. When the pressure in the U-tube is reduced, this bubble grows, and then violently collapses into the apex of the cone when the plate is raised. This collapse could generate sonoluminescence. Kosky and Henwood [12] utilized a similar geometry to study the pressures emitted as a result of vapour, rather than gas, bubble collapse.

The cone being transparent ($45 \pm 5\%$ of photons produced at the tip reaching the cone exterior), various optical instruments could be deployed to study the collapse, though not simultaneously because of their differing ambient lighting requirements. Strong illumination, provided by two synchronized stroboscopes of 20 μs flash duration, was required for video photography at 50 frames per second (f.p.s.) using a Hadland Photonics HSV. Operating at 25 f.p.s., a CCD camera (Photonic Science DS800) imaged not only the cone (weakly illuminated from behind by a flat beta light produced by painting phosphorous over a tritium source), but also the sonoluminescence. The CCD camera system produces each frame by interlacing two fields, each of 40 ms duration, each being 20 ms out of phase with the other. Therefore, in keeping with the 25 f.p.s. video recording, there is a frame every 40 ms, but in every such frame there is some information gathered over 60 ms. Each field integrates the light for only 18.4 ms (the remaining 1.6 ms being taken up by blanking filters). The persistence on the intensifier system is less than 3 ms for the exposures used here, and so will not affect the images presented here.

Both cameras (the HSV imaging the meniscus, and the DS800 recording the luminescence) were connected to a video recorder (Panasonic NV-FS88 HQ) using S-VHS videotape. The optical systems could not be deployed simultaneously because of their differing ambient light requirements. However, instrumentation to measure the pressure and indicate the level of the liquid could be deployed simultaneously with them. These instruments were mounted in the 62 mm tall polymethylmethacrylate (PMMA) extension to the shorter leg of the U-tube (Fig. 1). Within this extension, a pressure transducer (RS341-979), the centre of its 5.1 mm diameter placed 5 cm below the start of the cone, recorded the pressure fluctuations within the liquid close to the bubble. Such fluctuations measured by this 'low pressure transducer (LPT)' include the following: first, the pressure reduction before release of the plate (cross-checked with a pressure gauge fitted to the pumping train); second, the pressure wave which propagates through the U-tube in response to the opening of the plate; third, the rebound pressure pulses emitted by the bubble (the magnitude of the latter potentially being beyond the

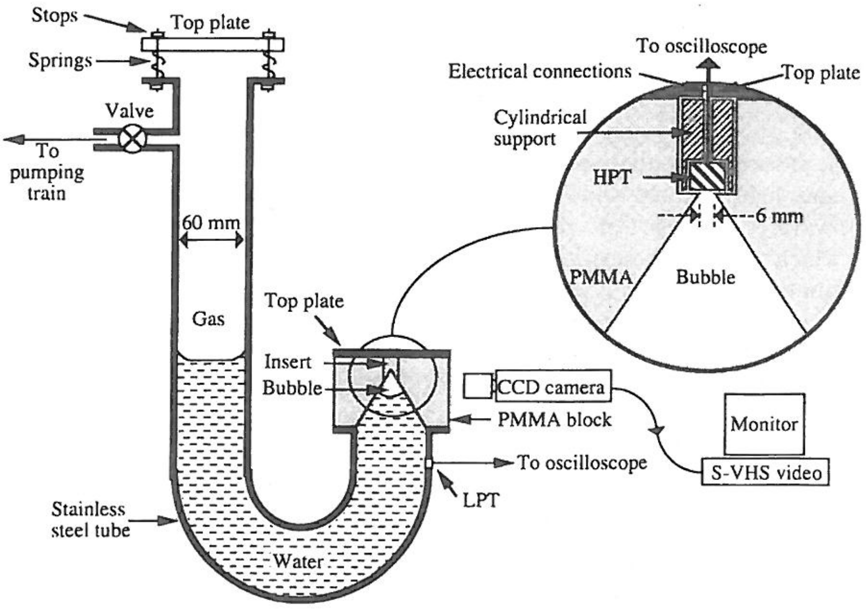

Fig. 1. Apparatus. The main figure shows the usual state of the apparatus (i.e. that used to take Figs. 3 and 5), with a polycarbonate insert, placed at the apex, completing the conical hollow in the polymethylmethacrylate (PMMA) block. The detail shows the arrangement used for Figs. 2 and 4, when the polycarbonate insert has been replaced by a mounting for the HPT.

0–0.21 MPa linear range of the LPT, outside which no calibration could be obtained from the manufacturers). The extension was removed when rebound pressures great enough to permanently affect its performance were planned. The characteristic response time of the LPT is 0.5 ms, indicating the limits of its temporal resolution.

The apex of the cone was formed from removable inserts (shown in Fig. 1), of 47 mm length and 25 mm outer diameter, containing a 30° half-angle conical space which, at its 13 mm diameter base, is commensurate with the aperture at the truncation of the main cone. The stresses at the cone tip resulting from the bubble collapse, be they gas pressures or the result of jets, are sufficient to damage perspex cone tips. Such an event is to be avoided, if only to remove the possibility that a contribution to the luminescence might otherwise be triboluminescent in origin. Therefore, for collapses capable of damaging PMMA cone tips, such as those reported in this study, polycarbonate cone tip inserts were used, which although tougher are less transparent.

Bubble collapses into conical polycarbonate inserts resemble to some extent the collapse of a solid angle segment of a spherical bubble. Such inserts were not employed exclusively in the studies. A second type of insert could be used to mount a pressure transducer (Keller PA-8, termed here high pressure transducer, HPT) which is calibrated up to 1000 bar, has a 13 mm diameter sensitive face, and a 30 kHz resonance. In mounting it, a 6 mm tall conical segment is placed between the transducer and the top of the truncated cone, so that the cone extends smoothly from its 60 mm diameter base to present a circular aperture of 6 mm diameter to the transducer (see detail in Fig. 1). Whilst this means that the whole of the sensitive area of the HPT is not presented to the bubble, so that the measurements of pressure are not absolute, not to reduce the aperture would fail to yield more accurate measurements. This is because during an energetic collapse of the type discussed here, the meniscus will travel up the conical walls, and the bubble will reach a size smaller than the circular aperture at the cone's truncation. At such a point two crucial events occur. First, some areas of the transducer exposed by the aperture will be in contact with gas, and some with liquid, and the resulting signal from the HPT will be some spatial average of the pressures in these. Second, the truncation of the cone is likely to strongly affect the collapse dynamics. The use of the 6 mm tall extension to the top of the cone prolongs the period before the effects set in.

The U-tube itself has the capacity to produce signals, quite apart from the oscillations of the bubble, when subjected to the impulse excitation resulting from the opening of the plate. To indicate what these signals might be, and allow them to be subsequently interpreted as non-bubble data when the collapse of a bubble is examined, the apparatus was tested with the water removed. Fig. 2 shows the signals consequently monitored by the LPT. Though the LPT might potentially respond to pressure signals in both the air and the solid structure into which it is mounted, the trace in Fig. 2 is dominated by airborne signals. The expected 1 atmosphere pressure step that results from the opening of the plate is followed by an oscillation, the fundamental frequency of which (111 Hz) corresponds to the first-harmonic of the semi-closed air-filled U-tube of length 73 cm. The HPT showed a slight response to the pressure step but no clear signal above the noise.

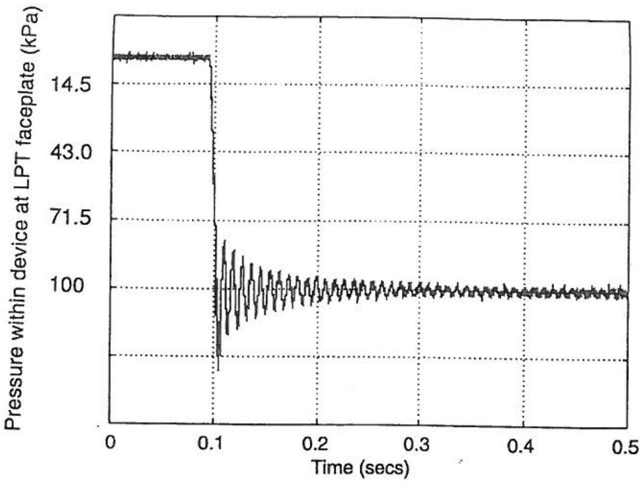

Fig. 2. Pressure records from the LPT when the vacuum is released in the absence of any water.

3. Results

Fig. 3 shows CCD pictures of the sonoluminescence from the cone tip. The cylindrical insert is clearly visible: its dark edges (spaced 25.1 mm apart, the diameter of the insert) are arrowed in Fig. 3(b), frame 1. Midway between these arrows the tip of the cone, hollowed out of this insert, is visible as a dark region, as it scatters the back-lighting away from the camera. Sonoluminescence is recognized as bright light sources within the cone, which appear on the video recording at the same time as the explosive sound that follows the release of the vacuum. All the following measurements from Fig. 3 are ±0.5 mm.

The luminescence generally appears in two main regions. Within the resolution of a frame, these may appear separately or together. In Fig. 3(a) the luminescence fills the cone from the apex to a distance of some 2 mm below it. In Fig. 3(b) no luminescence is detected at the cone tip, but rather occurs in an elliptical region some 4 mm × 2 mm, its centre being about 6 mm below the tip. The region is not symmetrical about the axis of the cone, but when it appeared in this form the location was asymmetric in the same direction (i.e. to the right

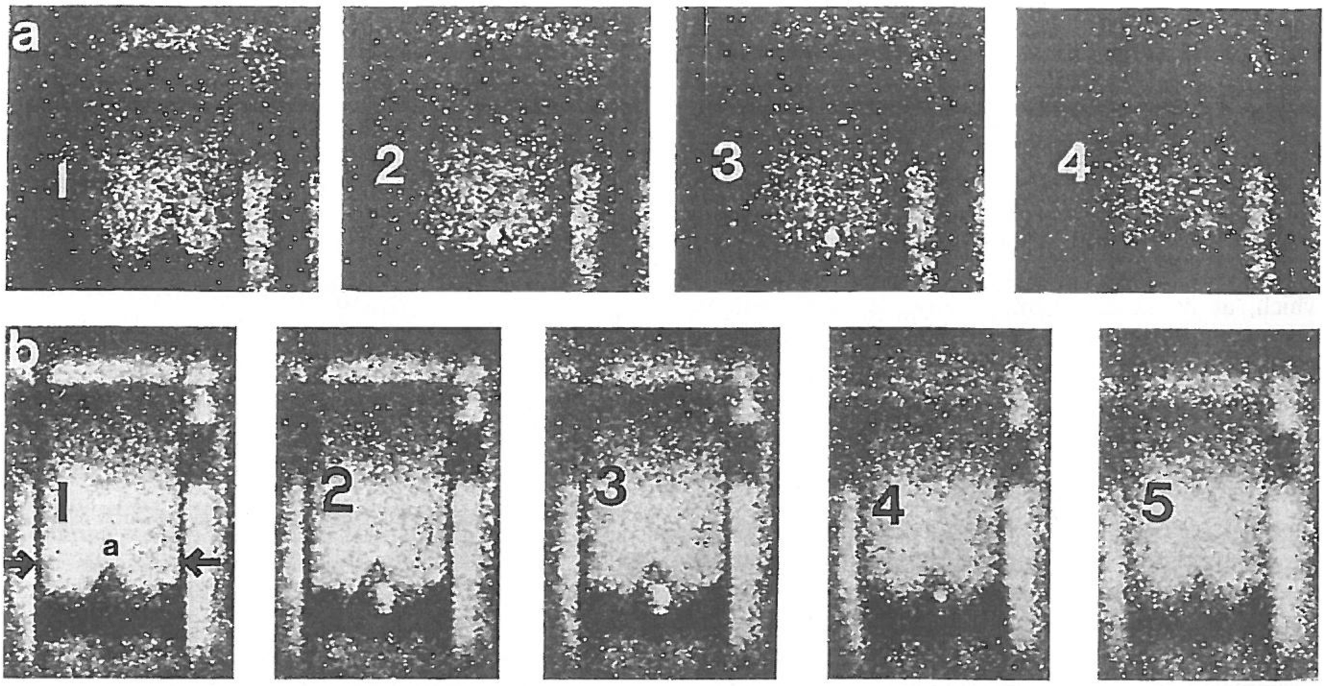

Fig. 3. Sonoluminescence imaged in the cone tip (interframe time = 40 ms, but each frame is interlaced as detailed in the text). Two collapses ('a' and 'b') are shown, with pre-collapse (i.e. fully expanded) bubble volumes of (a) 177 ml, and (b) 154 ml. In both cases the volume of water in the device was 1100 ml, and initial bubble volume was 0.065 ml. The boundaries of the insert, 25.1 mm apart, are arrowed in frame b1. Weak back-lighting is provided so that the apex of the cone (labelled with a black 'a' in frames a1 and b1) is shown in silhouette.

in this picture). This suggests that the source of the asymmetry, which defines the direction of the isotropy, lies in the apparatus itself. This region resembles[1] the shape of a rebounding cloud of bubble fragments shown in frame 3 of Fig. 5.

Whilst the luminescence is generated with a conical tip, addition of the HPT insert allows interpretation of the LPT output. Fig. 4 shows the simultaneous outputs from the HPT (lower trace) and LPT (upper trace) during a bubble collapse and rebound. The LPT records the pressure difference between its face and atmospheric. The first event is the initial pressure impulse caused by the release of the vacuum: the wave travels down the long leg and up the shorter leg of the U-tube, to be recorded as peak 'A' on the LPT output. This wave causes the bubble to collapse, the ambient gas pressure within it increasing until rebound occurs. This moment is indicated by the first peak on the HPT output (labelled '1'). The pressure wave emitted on rebound is detected by the LPT (peak 'B') a short time later, corresponding to the travel time in water between the LPT and the bubble meniscus roughly 11 cm away (i.e. close to the cone apex). The interval between peak positive pressures in '1' and 'B' is difficult to estimate, since both peaks contain structure, but appears to be 400 ± 300 μs. This gives an estimate for the spatially averaged sound speed between the LPT and the meniscus of between 1100 and 160 $\mathrm{m\,s^{-1}}$. These estimates are lower than the $\sim 1450\ \mathrm{m\,s^{-1}}$ sound speed [13] in bubble-free water at room temperature for an ambient pressure of 4.2 ± 2.7 kPa (estimated[2] from the base level of the LPT in Fig. 4).

Five subsequent peaks are apparent in the HPT trace (labelled '2'–'6'), corresponding to the bubble reaching minimum size, and shortly after each is the peak in the LPT corresponding to the passage of the pressure wave emitted on rebound (labelled 'C'–'G'). Subsequent peaks in the HPT are not discernible above the noise; however, the rebound pressure waves continue to pass over the LPT, giving rise to a series of peaks of gradually diminishing amplitude in the LPT output. The interval between LPT peaks corresponding to rebound emissions decreases with increasing time, it being around 12 ms between the first and second rebounds; whereas the low-amplitude peaks towards the end of Fig. 4 (i.e. after a time axis reading of about 0.08 s) occur at 7 ms intervals, their form becoming more sinusoidal. These might arise from two possible sources. They are most likely to be the pressure emissions corresponding to rebounds which occur sufficiently long after the initial collapse for the bubble wall oscillation to have been damped to a low-amplitude, more nearly linear form. A simple calculation

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[1] There is a resemblence, but the positions are not identical: a degree of difference between runs would be expected given that meniscus fragmentation is a result of an instability (see discussion of Fig. 5).

[2] This value corresponds to a measurement at the LPT, and includes a hydrostatic contribution (which will decrease closer to the cone) of 1078 Pa from 11 cm of bubble-free water.

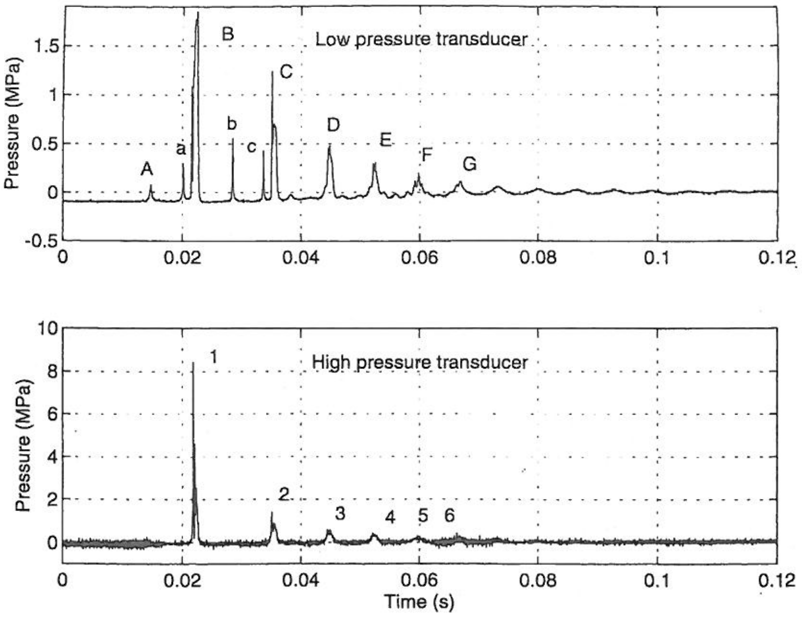

Fig. 4. The simultaneous records form the LPT (upper trace) and the HPT (lower trace). Prior to collapse, the bubble had grown from an initial volume of 0.38 ml to a maximum volume of 49 ml. The volume of liquid in the apparatus was 1100 ml.

disproves the second possible source, that of an 'organ-pipe' response in either the water- or air-column. For this experiment the former was 42 cm, corresponding to a fundamental frequency of about 1760 Hz, with a period of 0.6 ms, relatively independent of pressure. The air column is about 31 cm in length, corresponding to fundamental frequencies of about 270 Hz (i.e. a period of 3.7 ms) at 1 bar ambient pressure; and 190 Hz (i.e. a period of 5.3 ms) at an ambient pressure of 50 kPa (as can be seen from the LPT, the ambient pressure at this time oscillates around such values). All such harmonics therefore have periods less than the observed 7 ms, which suggests bubble oscillations as the source. The oscillations within peaks 'E', 'F' and 'G' are of order 0.3, 0.6 and 0.6 ms, respectively (all ±100 μs); the oscillations between peaks 'E' and 'F' are of period 1.9 ms. The source of such has yet to be conclusively identified.

A similar comment can be made regarding a number of very sharp peaks apparent in the LPT trace, which have no corresponding source in the HPT trace. One (labelled 'a') is clearly distinguishable between 'A' and 'B', and two (labelled 'b' and 'c') between 'B' and 'C'. As these did not appear in Fig. 2, their source is not in the mechanical operation of the device, such as the impact of the rising plate against the stopping bolts (which could in any event be cushioned). The hypothesis that peaks 'a', 'b' and 'c' are the result of reflecting and focusing gas shocks is not borne out, since there is no corresponding evidence for them in the HPT output. Since peak 'a' occurs prior to the initial rebound, the latter is clearly not its source. The only other source of a pressure wave is the initial peak 'A'. This will partially reflect off the meniscus, giving a pulse which travels back towards the plate opening. On reaching the planar meniscus in the long arm of the U-tube, the pulse reflects again, re-inverting back to a positive pulse, to again pass over the LPT. Pressure waves, emitted by rebounds (which, on passing over the LPT, give rise to signals at B, C, D, E, etc.) might also be expected to reflect at the planar meniscus in the long arm of the U-tube, and propagate back towards the cone, to then reflect off the bubble at its terminus. Whilst it is reasonable to expect such reflections to take place, the signal that the LPT will generate from the passage over it of such pressure pulses will depend on many factors, including: the response limitations of the LPT (in bandwidth, rise-time, linearity, and response to tension); non-linear propagation effects (which will affect the amplitude and temporal profile of a propagating pulse); and the fact that a wavefront which is almost planar as it passes over the LPT after propagation along the U-tube, may be distorted on reflection by the meniscus geometry and propagation in the cone, and by its effect on bubble fragments there (as Fig. 4 illustrates, a liquid tension can give rise to a much larger positive pressure pulse from bubble rebounds a short time later).

The hypothesis that peaks 'a', 'b' and 'c' arise through propagation paths involving reflection at the planar meniscus in the long leg of the U-tube can be examined against the interval between pulses. The measured interval between the detection of peaks 'A' and 'a' by the LPT is 5450±75 μs; between peaks 'B' and 'b' it is 6.3±0.4 ms; and between peaks 'b' and 'c' is 5200±50 μs. Though the ambient pressures and sound speed profile along the tube will not be constant, the similarity in these intervals might suggest it represents the time required to twice traverse the water column in the U-tube. However, given that it is 42 cm between the LPT and the meniscus in the long leg, the intervals are too great to be accounted for by the reduction in the speed of sound in pure water resulting from a decrease in ambient pressure [13] (though it varies both in time and position within the tube, the ambient pressure at the LPT is estimated from Fig. 4 to be 2 ± 2 kPa between 'A' and 'a'). Either the additional peaks are generated by rebound pressures from bubbles not attached to the HPT; or, recalling the sound speed estimates from the interval between peaks '1' and 'B', the sound speed has been reduced by bubble formation (through exsolution or, later, meniscus fragmentation).

When the HPT was replaced by a conical tip, the LPT trace still shows the same general form, indicating that the bubble rebounds for up to around 80 ms after the initial collapse. It is interesting to compare these timescales with those in Fig. 5, which shows a series of consecutive frames with interframe time of 20 ms. In frame 1, damage is clearly visible in the conical insert, the result of preceding impacts. In frame 2, the reflections in the cone are suddenly dimmed, indicating the rapid passage of the meniscus. By frame 3 a tight cluster of bubble fragments are visible, asymmetrically distributed near the apex. That liquid fills the cone to this level indicates that at least one collapse has occurred. By frame 4 this cluster of fragments has been ejected downwards (against buoyancy forces), whilst two more tight clouds of fragments are visible. These clouds migrate and disperse, eventually rising under buoyancy at the end of the event. Since Fig. 5 shows clouds of bubble fragments ejected from the rebounding bubble wall in frame 3, 20 ms after frame 2 shows the initial collapse, this suggests that despite extensive fragmentation some coherent gas pocket might remain at the apex tip to generate the characteristic HPT and LPT signals up to 80 ms after the initial collapse. A collective oscillation of fragments might be expected to affect the LPT only. Either a single body remains at the tip, its surface emitting fragments; or the whole bubble repeatedly fragments and coalesces at each rebound and collapse.

4. Discussion

The bubble collapses into the cone. It then behaves in such a way as to generate repeated pressure pulses

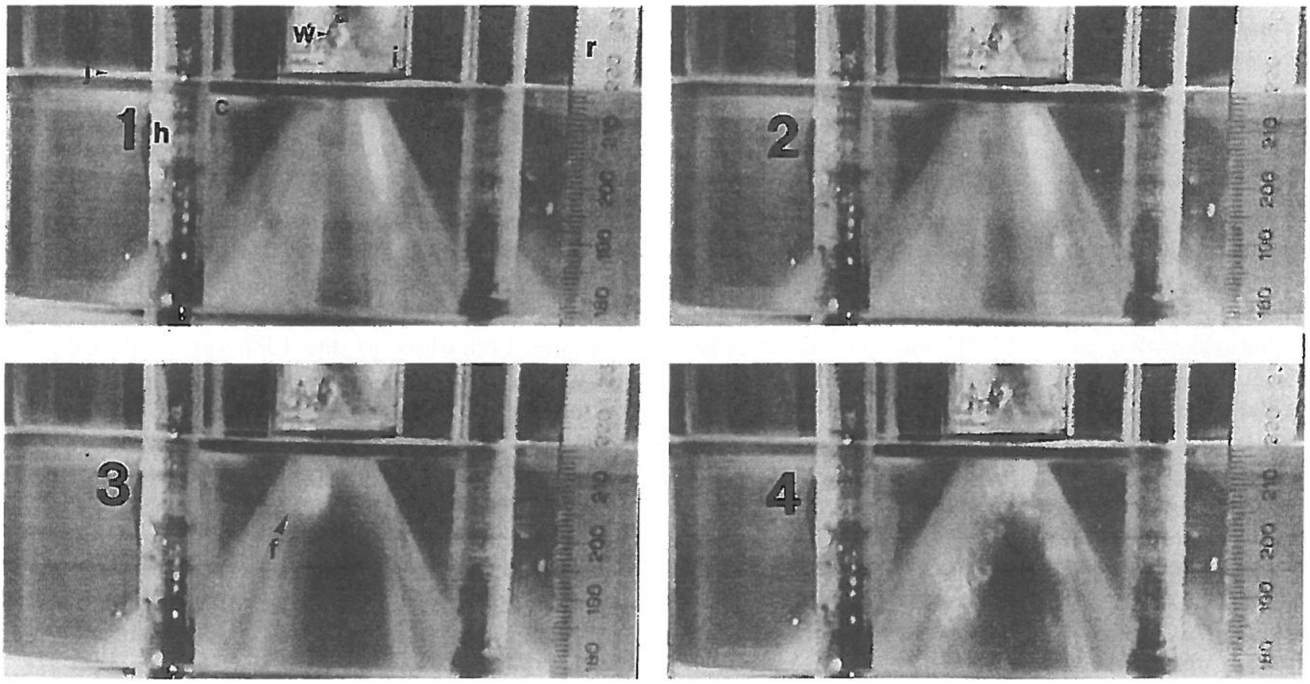

Fig. 5. Consecutive frames (filmed at 50 f.p.s.) of the bubble collapse. Frame 1, which has a millimeter ruler on the right (labelled 'r'), is filled by two PMMA blocks, the horizontal junction line between them being labelled 'j' and arrowed. Two holes, containing the bolts which hold the blocks together, are labelled 'h'. The upper block contains a cylindrical hole, its axis vertical, in which sits the insert (labelled 'i'). The insert comprises a cylinder of 25.1 mm diameter, containing the top section of the cone: the cone apex is labelled 'a'. Visible in the same region are water pockets (arrowed 'w') trapped between the outer edge of the insert and the surrounding perspex. The insert and its surrounding PMMA block sit on the main PMMA block (labelled 'c') out of which the remainder of the cone has been machined. The meniscus passes up the cone in the initial collapse during frame 2 (though visible on video, this is not clear in still photographs, as here). A cloud of fragments (arrowed 'f'), ejected from the rebounding bubble, is visible in frame 3. The cloud expands, and in frame 4 large bubbles (arrowed 'b') can be seen in it. Prior to collapse, the bubble had grown from an initial volume of 0.6 ml to a maximum volume of 145 ml, which it is sustaining during frame 1. The volume of liquid in the apparatus was 1100 ml.

both at the cone tip and 11 cm away in the U-tube, for several tens of ms after the initial collapse. The similarity in location between the cloud of fragments seen in Fig. 5, frame 3, and the luminescence in Fig. 3(b), frame 2, suggests that these clouds are capable of luminescence, and the transducer traces suggest that despite extensive fragmentation a gas pocket is present at the apex at the end of the collapses.

The geometry of the liquid is diverging within the cone, but non-diverging within the tube, where consequently an equivalent volume of liquid will contribute to a greater extent to the inertia associated with the motion of the bubble wall. It is therefore possible, within the limits imposed by the change of length of the liquid column in the tube, to choose the length of liquid in the tube to mimic the inertia that would be found in the spherical collapse of a bubble in an infinite body of water. Furthermore, by increasing the length of liquid in the tube, the inertia associated with the collapse is increased, simulating to a certain extent bubble collapse within a more dense liquid.

5. Conclusions

This paper introduces and performs initial charaterization of a device which produces sonoluminescence from the collapse of, initially, a single bubble. Though photographic evidence suggests that bubble fragments are ejected from the bubble wall on rebound, comparison with pressure measurements within the liquid suggest that even at the end of the collapse a bubble remains at the cone tip, ready to repeat the cycle. Propagation times of the pressure pulses emitted on rebound can be used to investigate the local sound speed.

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