Research Notes

Transient excitation of insonated bubbles

T.G. Leighton

Cavendish Laboratory, Madingley Road, Cambridge
CB3 OHE, UK

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When a previously non-oscillating bubble is first insonated, its radial motion can contain transients. Before the oscillation settles down to the steady-state, these transients may enhance the violence of the volume pulsations. The temperature attained within the bubble can be much greater than that reached during the steady-state motion. This phenomenon may contribute to the increased cavitational effects (such as sonoluminescence or ultrasonic bioeffects) observed when ultrasound is pulsed, and when the transducer and liquid sample are in relative motion.

Keywords: sonoluminescence; cavitation; bubble dynamics; bioeffects

When liquids are subjected to ultrasound, bubbles in them may be forced into oscillation, and so sonoluminescence. It has been noted that in certain circumstances sonoluminescent activity may be enhanced. Leighton *et al.*[1] insonated a sample of aerated water with 1 MHz continuous-wave ultrasound at 3 W cm^{-2} acoustic intensity, using a Therasonic 1030 (Electro-Medical Supplies) transducer. Flashes of sonoluminescence were seen when the transducer was rocked (mimicking its movement when it is massaged on the patient's skin during physiotherapeutic treatment). During this activity, the acoustic beam is scanned across the bubble field. Sonoluminescent flashes were also seen each time the Therasonic unit was switched on.

In this study, an explanation for this is proposed and tested. The common factor in the two situations described above is that previously non-oscillating bubbles are subjected to insonation. Before their radial oscillations settle down to the steady-state motion, short-lived transients in the function may enhance the violence of some collapses. The minimum volume reached by the bubble during an oscillation is smaller than that expected from the steady-state motion, and so the temperature reached by the gas adiabatically compressed inside the bubble is consequently higher. This will result in an increased intensity of sonoluminescence. Therefore, we observe the flashes of luminescence in the situations outlined above.

Materials and methods

The RPNNP (or Rayleigh–Plesset) equation describes the motion of a spherical gas bubble in an incompressible liquid[2]. This equation is commonly used to predict the steady-state oscillations of bubbles[3]. In this study it has been solved numerically to include the effect of the transients in the function which occur when a bubble is first insonated. At time $t < 0$, the bubble is at equilibrium size and the bubble wall is stationary. Insonation starts at time $t = 0$. The bubble radius, wall velocity, and the temperature and pressure of the gas contained inside the bubble are plotted against a common time axis in *Figures 1* and *2*. It is assumed that the bubble maintains a spherical shape throughout the motion (assumed to be adiabatic). The plots are for air bubbles in water at normal atmospheric pressure. The bubbles represented in *Figures 1* and *2* are of a size just below that resonant with the acoustic field. They undergo stable cavitation. Bubbles of much less than resonance size in a 10 kHz sound field are seen to undergo unstable cavitation (also known as transient cavitation, a term not employed in this study to avoid confusion with the transients that occur in the function of the radial motion). For these

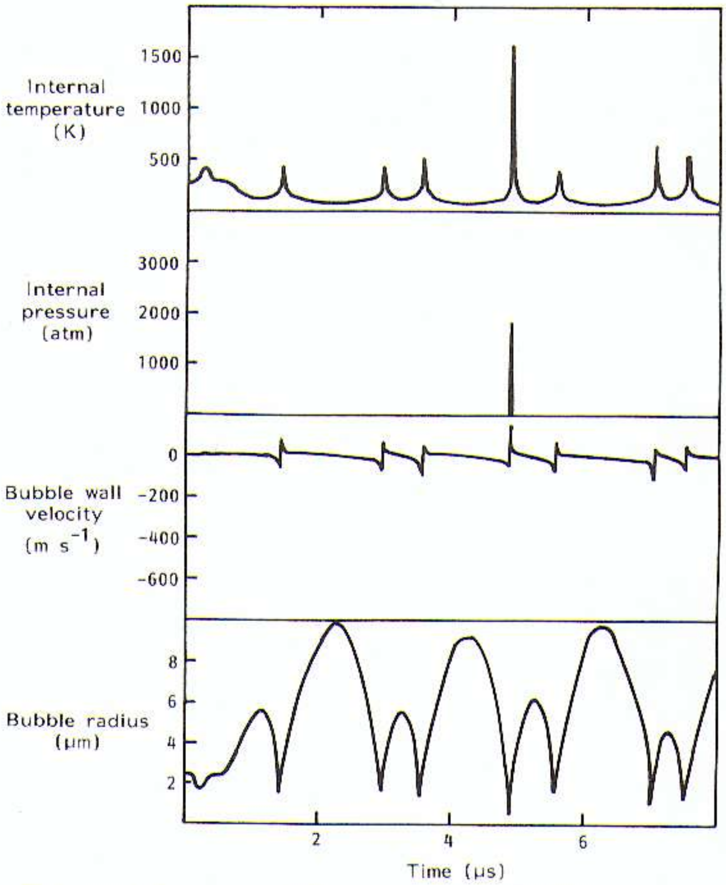

Figure 1 Computer plot of the response of an air bubble in water at a liquid temperature of 20°C. Equilibrium bubble radius is 2.5 μm; the acoustic pressure amplitude is 2.4 atm. at a frequency of 1 MHz. Insonation begins at time $t = 0$ (the bubble is at rest for all times $t < 0$). Bubble radius, wall velocity, and the pressure and temperature of the gas within the bubble are plotted against a common time axis

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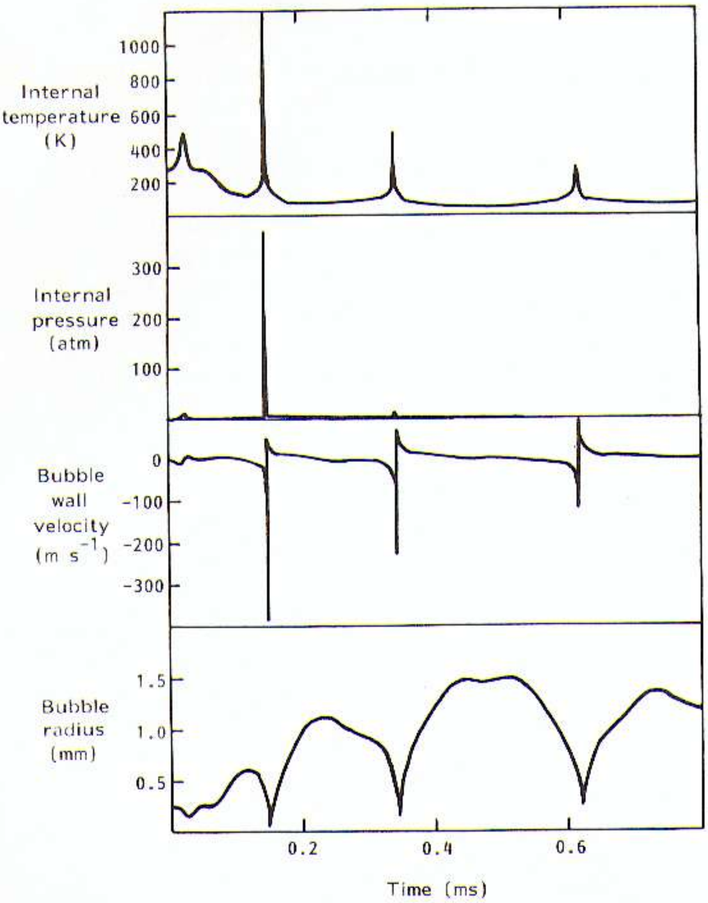

Figure 2 Computer plot of the response of an air bubble in water at a liquid temperature of 20°C. The equilibrium bubble radius is 0.25 mm; the acoustic pressure amplitude is 2.4 atm. at a frequency of 10 kHz. Insonation begins at time $t=0$ (the bubble is at rest for all times $t<0$). Bubble radius, wall velocity, and the pressure and temperature of the gas within the bubble are plotted against a common time axis

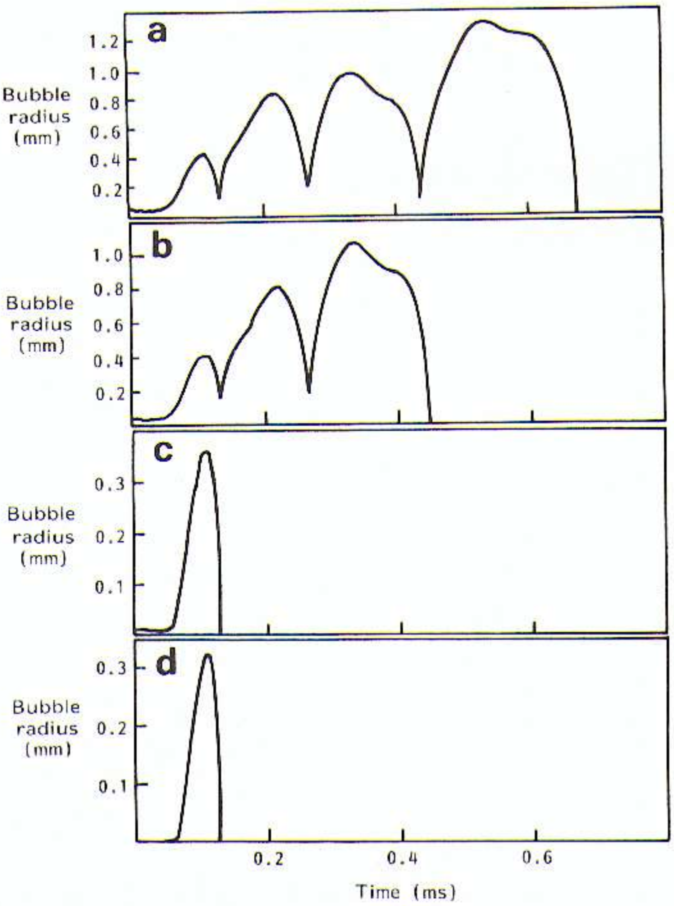

Figure 3 Computer plots of the response of four isolated air bubbles in water at a liquid temperature of 20°C. The acoustic pressure amplitude is 2.4 atm. at a frequency of 10 kHz. Insonation begins at time $t=0$ (the bubbles are at rest for all times $t<0$). Equilibrium bubble radii: (a) 60 μm; (b) 50 μm; (c) 10 μm; (d) 1 μm

bubbles, the radial motion alone is plotted as a function of time (*Figure 3*).

The effects of transient excitation on sonoluminescence were observed experimentally in a 10 kHz sound field using an EMI 9757B photomultiplier tube. The sound was generated by a piezoceramic cylinder (diameter 7 cm) and was focussed to produce a presure antinode on the axis of the cylinder (which was filled with water). The photomultiplier views vertically along this axis, from above. *Figures 4a* and *b* show the photomultiplier output plotted on a common time axis with the simultaneous hydrophone trace (taken with a Bruel and Kjaer type 8103).

Results

Numerical solutions

Bubbles of near-resonance size. If ν is the insonation frequency, then the resonant bubble radius, R_r (the natural frequency of oscillation of an unforced bubble), for an air-filled bubble in water is given[4] by the formula $\nu R_r \approx 3\ \mathrm{Hz\ m^{-1}}$. Both *Figures 1* and *2* show the response of a bubble of just less than resonance size. *Figure 1* shows the response of a bubble of equilibrium radius 2.5 μm to a 1 MHz field with a 2.4 atm. pressure amplitude (1 atm. = 10^5 Pa). The transient excitation causes the internal temperature to exceed 1500 K, approximately four times that reached during the steady-state motion. A similar effect is seen in the response of all bubbles of just less than resonance size over the whole frequency range investigated, from 1 MHz to 10 kHz. *Figure 2* illustrates the response of a bubble of 0.25 mm equilibrium radius to an acoustic field of 2.4 atm. amplitude and of frequency 10 kHz. This bubble is undergoing stable cavitation, which is similarly enhanced by transient excitation in that the internal temperature peaks at about 1200 K during one oscillation.

The numerical analysis used here does not incorporate the effects of heat conduction from the bubble, since the original RPNNP equation does not include thermal damping. Hickling[5] and Fujikawa and Akamatsu[6] both investigated the conduction of heat from bubbles and its effect on short-lived, impulsive phenomena, though neither studied stable acoustic cavitation. Hickling examined unstable cavitation, whilst Fujikawa and Akamatsu theoretically modelled how heat and mass transfer across the bubble wall affect the pressure wave generated when a bubble is collapsed by a water shock tube (an experiment performed by Fujikawa and Akamatsu[7]). However, both concluded that heat conduction is negligible for nitrogen bubbles of an initial size of about 10^{-4} m. For example, both the internal bubble temperature (Hickling[5]) and the impulsive pressure generated in the water (Fujikawa and Akamatsu[6]) are reduced by only about 5% if heat

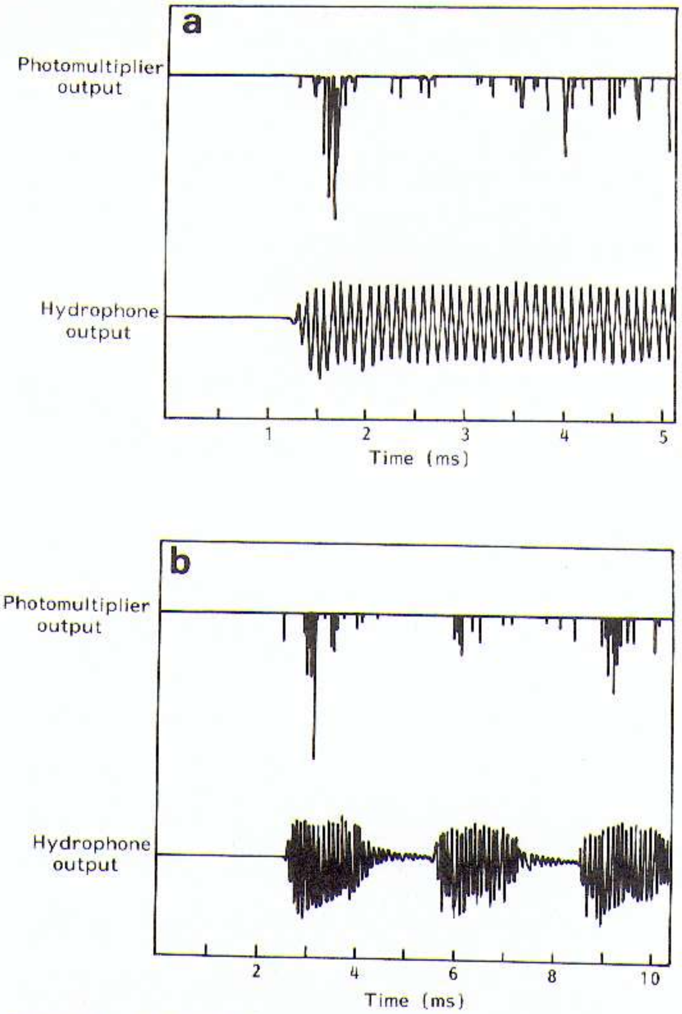

Figure 4 (a) Simultaneous photomultiplier and hydrophone outputs are shown on a common time axis for the insonation of water at 10 kHz, with 2.4 ± 0.1 atm. pressure amplitude, at the start of continuous-wave insonation. (b) Simultaneous photomultiplier and hydrophone outputs are shown on a common time axis for the insonation of water at 10 kHz, with 2.4 ± 0.1 atm. pressure amplitude. Three pulses of sound are used: both the pulse length and the separation are equal to 16 acoustic cycles

conduction is allowed for. Both also found that as the initial bubble size decreased, the discrepancies become greater. Hickling found that nitrogen bubbles of initial radii of 10 μm attain only about 50% of the internal temperatures that the calculations that ignore heat conduction reach; Fujikawa and Akamatsu found that the discrepancy in the impulsive pressure generated increased as the collapse progressed, reaching extremes where it is reduced to about 30% of the magnitude calculated without heat conduction.

Though the situation is not identical here, these findings suggest that the transient excitation of near-resonance sized bubbles at 10 kHz will not be much affected by heat conduction. However, at 1 MHz it may be so. Estimates can be made to test this. The thermal diffusion length in time t from a bubble (Jost[8]) is about $(D_t t)^{1/2}$, where D_t is the thermal diffusivity of water (where $D_t = 1.43 \times 10^{-7}$ m^2 s^{-1}). From *Figure 2*, the thermal spike induced by transient excitation can be estimated to endure for 2 μs; therefore, the thermal diffusion length for this will be about 5×10^{-7} m. This is 0.25% of the equilibrium bubble radius, thus, as expected from the results of Hickling and Fujikawa and Akamatsu, the effects of thermal conduction will not greatly affect the temperature attained. The thermal spike in *Figure 1* exists for about 10^{-8} s; the thermal diffusion length is therefore 10^{-8} m, which corresponds to 0.4% of the equilibrium bubble radius. So here also the effects of thermal diffusion are not likely to be great. Therefore, at both frequencies, the conduction of heat from the bubble could not extinguish the thermal spike, and so flashes of sonoluminescence resulting from transient excitation will result.

Bubbles of much less than resonance size. A second effect is seen at 10 kHz. As the equilibrium bubble size decreases, the cavitation changes from stable to unstable. In the sound field with 2.4 atm. pressure amplitude, numerical solution (not included here) shows bubbles with equilibrium radii of 65 μm undergoing stable cavitation. However, between 65 and 60 μm, the motion changes to unstable. *Figure 3a* shows the oscillation of a bubble of 60 μm equilibrium radius. After a finite number of oscillations, an unstable collapse occurs. *Figures 3b, c* and *d* show the radial motions of bubbles of equilibrium radius 50, 10 and 1 μm, respectively. As the initial size becomes less, the bubble performs fewer oscillations before becoming unstable. Both bubbles in *Figures 3c* and *d* reach a similar maximum size (about 0.3 mm) before collapsing (as expected from the work of Apfel[9]).

Preliminary investigations at 10 kHz suggest that as the acoustic pressure amplitude is increased, the critical equilibrium bubble radius above which the cavitation is stable, and below which it is unstable, increases. For example, when the amplitude is 2.7 atm., the crossover occurs at a radius of about 100 μm.

In calculating the bubble internal pressure and temperature in *Figures 1* and *2*, adiabatic conditions were assumed. Though acceptable for cases of stable cavitation, it would not be so for unstable cavitation, since most authors agree that the growth phase in an unstable collapse is isothermal[2].

An estimate of the temperature reached during the unstable collapse of the bubbles in *Figure 3* can be made. The exact solution cannot be found, because as the bubble size reduces indefinitely, the internal temperature will increase without limit. In practice, surface instabilities set in at some point, causing bubble break-up. Plesset and Mitchell[10] found that during unstable cavitation, instabilities become violent when the bubble radius is one-tenth of the maximum radius (attained at the start of the collapse phase). If the growth phase in unstable cavitation is assumed to be isothermal, then the instabilities will set in when the bubble radius is one-tenth of the maximum radius reached during growth. Having, therefore, an upper limit for the minimum radius reached during collapse, and assuming an isothermal growth phase and an adiabatic collapse, the temperature reached during each collapse can be found. At the start of the collapse, the bubble temperature is that of the surrounding liquid, since the growth was isothermal. The collapse occurs adiabatically until the radius is about one-tenth of the maximum, say. Therefore, the bubble temperature at the end of the collapse will be about $10^{3(\gamma-1)} \times 300$ K = 4755 K. This rough estimate shows that these unstable collapses may well produce sonoluminscence directly. (Even were these temperatures not great enough to cause luminescence directly, the unstable collapses could encourage sonoluminescence some time later, since the bubble fragments from these collapses[11] may grow by rectified diffusion to a size large enough to sonoluminesce.) These unstable collapses have been photographed at 8000 frames per second using a Hadland Hyspeed camera. The radial motion is as predicted by these numerical solutions.

Thus, the enhanced stable cavitation of near-resonant sized bubbles, and the unstable collapses of bubbles with radii much smaller than resonance, may both result in flashes of sonoluminescence through transient excitation. Detection of these flashes is discussed in the following section.

Detection of transient excitation by photomultiplication

Transient excitation causes enhanced collapse during stable cavitation in bubbles of just less than resonance size, and unstable collapse in bubbles which are very much smaller than resonance. These would result in flashes of sonoluminescence. *Figure 4a* shows the photomultiplier output and the simultaneous hydrophone trace taken when insonation of water at 10 kHz begins (the acoustic pressure amplitude is again 2.4 atm.). Enhanced sonoluminescent flashes are detected by the photomultiplier within a few acoustic cycles of the start of insonation.

Another situation in which previously quiescent bubbles may begin to oscillate is when the sound is pulsed. If the off-time is of sufficient length, any bubble oscillations will be damped out. At the start of the next sound pulse, transient excitation may occur. In *Figure 4b*, three acoustic pulses (each 16 acoustic cycles in length and with an off-time of the same length) of a 10 kHz fundamental were used to insonate water. The photomultiplier and hydrophone outputs are recorded simultaneously, as in *Figure 4a*. The flashes of sonoluminescence predicted by the transient excitation theory are clearly visible near the start of each pulse. Those for the first pulse are the most intense since the steady-state motion will not be completely damped out during these off-times, thus the transients will have a less violent effect for the subsequent pulses.

Conclusions

Transient excitation can cause flashes of sonoluminescence by: 1, enhancing the stable cavitation of bubbles of just less than resonance size; 2, causing the unstable collapse of bubbles which are much smaller than resonance. Transient excitation can be brought about: 1, by scanning an acoustic beam across a bubble field; 2, at the start of insonation (particularly when the sound is pulsed).

Transient excitation may contribute to some examples of the enhancement of cavitational effects seen by other workers. Pickworth *et al.*[12] observed enhancement of sonoluminescence on pulsing 1 MHz therapeutic ultrasound. Hill *et al.*[13] increased acoustically-induced DNA degradation and KI decomposition by pulsing a 1 MHz sound field. Ciaravino *et al.*[14] and Flynn and Church[15], also working at 1 MHz, observed an increase in iodine release for certain pulsing regimes. Church amd Miller[16] and Morton *et al.*[17] have produced increased cavitational effects by rotating biological material in an acoustic beam (equivalent to rocking the transducer, as described above). A similar result was observed by Sacks *et al.*[18] on mammalian cell lysis. The close similarity between the systems used in these cases and the ones employed above to produce transient excitation of bubbles, suggests that the latter may have been a contributory factor in the enhancement of the cavitational effects quoted.

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