A new mechanism for sonoluminescence

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(Received 12 June 1996; accepted for publication 11 November 1996)

It is argued that a pulsating acoustically levitated bubble cannot possibly maintain a spherical shape. A jet forms during compression, and the sound amplitude such that the jet first strikes the other side of the bubble with sufficient energy is hypothesized to be the threshold for sonoluminescence. It is proposed that the connection between jet impact and light emission is a fracturing of the liquid that cannot flow during the extremely short time scale over which pressure is applied. With this hypothesis, sonoluminescence would therefore be a manifestation of the non-Newtonian nature of water and other simple liquids when stressed with sufficient intensity and rapidity. © *1997 Acoustical Society of America.* [S0001-4966(97)03104-4]

PACS numbers: 43.35.Ei, 78.60.Mq [HEB]

After its original discovery by Gaitan and Crum,[1] the fascinating phenomenon of stable, single-bubble sonoluminescence has attracted considerable attention for its unexpected and unique features.[2–11] Currently, the accepted working hypothesis as to its origin is the generation of a shock wave in the gas, its focusing at the center of the bubble, and the consequent formation of a plasma.[12] In my view this hypothesis faces serious difficulties both on a theoretical and an experimental level. The purpose of this note is to substantiate this statement and to suggest a possible alternative explanation of the phenomenon.

Most of the work carried out to date assumes that the bubble maintains a more or less spherical shape in the course of the oscillations.[13] A consideration of the mechanism responsible for the trapping of the bubble in the sound field uncovers some problems with this picture. Since bubbles tend to move in the direction opposite to the local pressure gradient $\nabla P(\mathbf{x},t)$, in a sound field a bubble drifts toward the pressure minimum (i.e., the antinode) when the pressure falls and toward the pressure node when it rises. If the bubble is driven below resonance, its volume v expands when the pressure falls so that the force $-v\nabla P$ is greater in magnitude during the expansion than during the contraction phase. Thus, the bubble executes a periodic translational motion in which the upward displacement in the compressed state, under the action of gravity and of the acoustic pressure gradient, is exactly equal to the pressure-gradient-induced downward displacement in the expanded state against gravity.

Observation as well as computation and a simple ''thought experiment'' all show that translation causes the formation inside the bubble of a jet oriented in the direction in which the bubble moves during the compression half-cycle. Consider a spherical bubble with negligible internal pressure that is released with a certain initial translational velocity. The bubble will start collapsing and its translational velocity U to increase due to the conservation of the liquid impulse $\frac{1}{2}v\rho U$, where ρ is the liquid density. It was pointed out a long time ago by Benjamin and Ellis[14] that, if the bubble were to collapse spherically and (in the idealized case of an empty bubble) reduce to a vanishing point, the impulse of the system could not be conserved. Instead a jet forms, directed in the same direction as the translational velocity, that will ultimately span the bubble and give rise to a toroidal vortex that conserves the total impulse. The first photographic evidence was provided by Benjamin and Ellis in Fig. 2 of their paper. A numerical simulation, generated by a standard boundary integral method documented elsewhere (see, e.g., Ref. 15), is shown in Fig. 1. Results of this type have been available in the literature for many years (see, e.g., Ref. 16 and references therein).

The point of this argument is to demonstrate that, during the collapse of a translating bubble, fluid dynamic conditions must prevail that tend to promote the formation of a jet. In a sound field, whether this jet traverses the entire bubble or has time only to develop to the embryonic stage of a dimple depends on the amplitudes of the volume pulsation and of the translational motion, i.e., ultimately, on the sound level.

Figure 2 shows a sequence of bubble shapes (computed by the same method used for the previous figure) in which the effects of surface tension, gravity, and spatial variation of the acoustic field are included. The latter is specified to be $P(z,t) = P_\infty + P_a \sin(\omega t)\cos(\omega z/c+\phi)$, with $P_\infty = 1$ bar, $P_a = 1.35$ bar, $\omega/2\pi = 26.5$ kHz, $c = 1{,}500$ m/s, $\phi = 0.58$ rad, and the time unit is 1.1 μs. Gravity acts downward along the z axis, the liquid is water, and the gas behaves isothermally. Initially the bubble is at rest with its center at $z = 0$ and an equilibrium radius of 4.5 μm. A similar calculation, but extending over more than one cycle and demonstrating the stabilizing effect of surface stretching during the expansion phase, may be found in Ref. 17. Although none of these examples exactly simulates the situation prevailing during sonoluminescence (the motion is neither steady nor periodic, viscous effects are ignored, the bubble internal pressure is calculated crudely, etc.), the point here is to substantiate the preceding conclusion that a jet forms in the direction of the bubble translational velocity during the compression phase. Another aspect worthy of notice in Fig. 2 (especially in connection with results recently published in Ref. 18) is the nearly perfect spherical shape of the bubble until less than 10 ns before the jet strikes the other side.[19]

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 0001-4966/97/101(4)/2003/5/$10.00

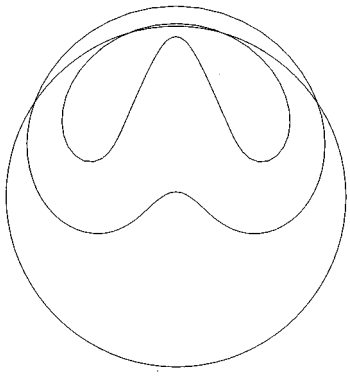

FIG. 1. Successive shapes of an axisymmetric translating and collapsing bubble in an unbounded, inviscid, incompressible liquid. The initial upward velocity of translation equals $0.6\times\sqrt{\Delta P/\rho}$, where ΔP is the difference (assumed constant) between the internal and external pressures and ρ is the liquid density. The successive shapes shown are at times 0, 0.6, and 0.86 in units of $R_0\sqrt{\rho/\Delta P}$. For $\Delta P=1$ bar and the density of water, with $R_0=10$ μm, this equals 1 μs. Surface tension is neglected. Note the jet directed in the same direction as the translational velocity. The jet becomes thinner with increasing translational velocity.

If one accepts then that, in the conditions prevailing during sonoluminescence, a bubble translates up and down with a jet that forms during collapse and disappears during expansion due to the stretching of the interface, and that the jet extends more and more into the bubble as the translation and pulsation amplitudes increase, it is then obvious that there must be a threshold value of the sound pressure (depending on bubble radius and other parameters) such that the jet just makes it across the bubble. I propose that this is the threshold at which sonoluminescence first sets in (provided the impact velocity is sufficiently large).

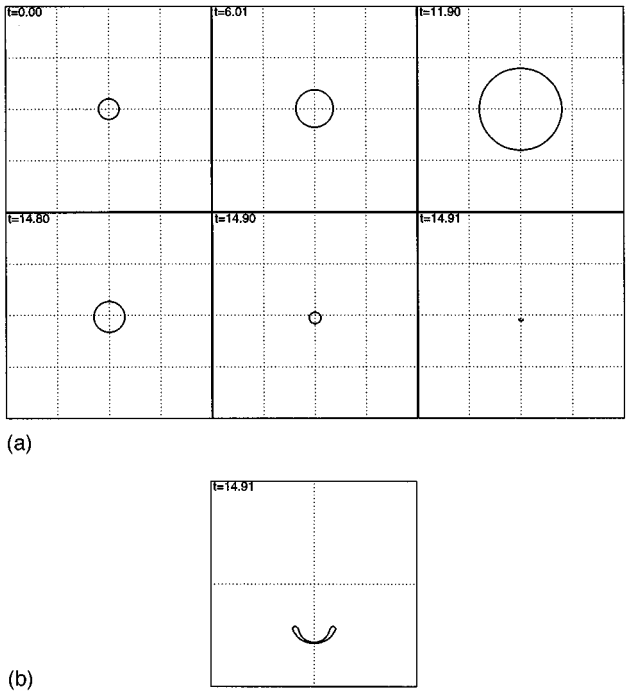

FIG. 2. Successive shapes of a 4.5-μm radius bubble in water placed in a pressure gradient in an unbounded liquid and subject to a pressure field oscillating at 26.5 kHz. The flow is assumed to be axisymmetric, irrotational, and incompressible, and the bubble initial velocity vanishes. Surface tension effects are accounted for. Times are in units of 1.1 μs. Figure (b) is an enlargement of the last frame of (a).

Before discussing the point further, let me address what in my view is a serious weakness of the shock wave hypothesis, namely that ''it explains too much.'' For clarity, let us separate two aspects of the experimental evidence. On the one hand there is the fact that light is emitted at all. On the other is the issue of stability and repeatability. The key to the second aspect has been correctly identified, I believe, in the balance that must prevail among the processes responsible for acquisition and loss of dissolved gas by the bubble.[9] But if shock waves are the light-emission mechanism, then why is luminescence (but not, necessarily, stability) not observed more commonly than it actually is? In other words, with a shock wave mechanism, one would expect to see quite easily light on and off for one or a few cycles as bubbles translate and evolve in a sound field under a variety of conditions, pressure amplitudes, etc. Yet this does not seem to happen in a normal cavitation field at weak to moderate amplitudes, but only at the very elevated amplitudes characteristic of the earlier work on sonoluminescence (see, e.g., Ref. 20). In the framework of the shock wave model, this lack of emission is explained with the hypothesis that translating bubbles distort too much to provide the necessary focusing. Yet, some bubbles ''can be seen to emit light as they rise from the nichrome wire,''[9] which seems to be in contrast with this explanation. As pointed out in Ref. 11, the shock wave hypothesis has another weakness, namely that it has difficulties accounting for the extreme sensitivity of the phenomenon to temperature, the nature of the liquid, and that of the gas. This argument may be countered by pointing out that changing conditions changes the nature of the gas in the bubble, a point that could be settled by experiments on the effect of similar changes on shock wave luminescence in standard shock-tube experiments. A further difficulty with the shock wave hypothesis is the recently reported observation of sonoluminescence with gases, such as ethane, having an adiabatic index close to 1.[8,9]

The argument often heard to oppose the notion that jets straddle the bubble is the observation that the bubble maintains its integrity for a very long time. Yet, there are at least two pieces of experimental evidence showing that jets do not necessarily destroy the bubble. The first one is the observation by Crum of a large bubble pulsating near a wall on a vibrating table.[21] At every cycle a jet forms that bridges the bubble during compression and is torn apart during expansion. While the parameter values for this experiment were vastly different from those prevailing during sonoluminescence, the fact remains that there is no necessary connection between jet formation and bubble disintegration. A more cogent observation was reported over 20 years ago by Lauterborn and Bolle, who produced a bubble near a solid wall with a focused laser flash and filmed the event at 75 000 frames per second.[22] In their beautiful Fig. 2 the bubble is observed to grow first and then collapse. After undergoing a strong distortion near the point of minimum volume, it rebounds (due to inflow of dissolved gas during the previous expansion), maintaining its integrity and a very nearly

spherical shape, except for a curious ''stem'' on the side near the wall. This feature is produced by the impact of a microjet that has traversed the bubble and that is visible in its interior while being stretched by the rebound.

Most interestingly, the stem of the bubble in Lauterborn and Bolle's sequence is eventually seen to undergo a separate collapse, leaving behind a number of minute microbubbles. This observation is of particular relevance to the present discussion because it offers a likely explanation of the observed decrease of the equilibrium size of the bubble corresponding to the onset of sonoluminescence, and it also explains the gas loss of about 0.01% per cycle that is estimated from diffusion theory.[9] In the words of the authors of Ref. 9, ''the process that leads to this [gas] ejection is the key to SL in a single bubble.'' The hypothesis of a connection between this ''anomalous'' mass loss and the stem collapse is obvious. The shedding of microbubbles has also been offered as an explanation for ''phase glitches'' observed with argon bubbles.[8] Another piece of evidence in favor of the above scenario is that, contrary to the nonluminescing case, a luminescing bubble is observed to bounce around its minimum radius less than a nonluminescing one.[9] This behavior may be due to the fact that the jet threading the bubble represents a significant energy loss that inhibits the bouncing.[23]

In summary, my hypothesis so far is the following: *The sonoluminescence lower threshold coincides with the pressure amplitude at which the jet developing during the collapse phase impacts (with sufficient energy) the opposite side of the bubble.* The upper threshold is due to the total disruption of the integrity of the bubble when the impact becomes too violent.

If one rejects the shock wave mechanism, what is the origin of the light emission? How is it connected with the jet striking the opposite side of the bubble? At this point I must leave what I believe are rather solid grounds and venture into speculation. The mechanism I propose is *fracto-luminescence,* the emission of light associated with the ''fracture'' of the liquid.[24]

It is a documented fact that, when some solid materials (including ice and Wint-O-Green Lifesavers® candy) fracture, light emission is observed (see, e.g., Refs. 25–30). This effect can be due to several factors, the relative importance of which has not been entirely sorted out. It is possible that the material behind the crack tip is left in a highly excited state due to nonadiabatic processes associated with bond breaking,[29,30] or, stated differently, that plastic deformation produces a strong thermal excitation. Other possibilities are defect production and electron capture, electron bombardment of the freshly created surface, and charge separation during crack growth followed by microdischarge.[29] The last mechanism is supported by the presence of spectral lines of the ambient gas in some (but definitely not all) experiments.[25,26] Interestingly, the spectra observed by Chapman and Walton[25] and others during the fracture of quartz were continuous, with no spectral lines, and resembled a blackbody spectrum, qualitatively very similar to single-bubble sonoluminescence spectra.[6]

While there is no actual proof that something resembling fracture does indeed occur in sonoluminescence, a series of considerations may make it at least plausible.

The first question is whether the very process of fracture, as opposed to flow, is actually possible. The following quotation from a section entitled ''The rigidity of liquids'' in Ref. 31 is relevant here: ''If the rate of shear is sufficiently great there may not be time for the [liquid] molecules to advance to a neighbouring site. In this case the liquid will not show viscous flow, but will show a finite elastic rigidity.'' An example is silicone putty, which fractures in a brittle fashion under rapid stretching, but flows in a viscous manner at low rates. Continuing with the quotation ''For simple liquids the rates of shear for this [i.e., rigidity] to occur are enormous...'' but not infinite. In other words, any liquid will exhibit a non-Newtonian behavior over sufficiently short time scales. Water is particularly likely to exhibit such ''rigidity'' due to the strong hydrogen bonding that confers to the liquid a structure bearing unusual similarities to the crystalline state. If stresses are applied too fast for relaxation, i.e., flow, to take place, the hydrogen bonds remain ''frozen.'' This peculiarity of water (together with another one mentioned below) may explain why ''water is by far the most friendly fluid for SL.''[8] An estimate of the necessary rate may be found if one assumes that the relevant time scale cannot be shorter than the inverse of the vibrational frequency of water molecules, which is of the order of 10^{13} Hz. If the microjet hits the other side of the bubble with a velocity U, the rise time of the overpressure can be assumed to be a mean intermolecular distance d divided by U. This estimate assumes that the most superficial layer of molecules is instantaneously accelerated to the velocity U and has to travel a distance d to compress the next layer. Assuming $d \sim 1$ Å, we have $d/U \sim 10^{-13}$ s for $U \sim 1{,}000$ m/s which is of the order of the speed of sound in the liquid (as expected), and of the estimated radial bubble velocity and therefore of the jet's velocity as well.

The duration of the tremendous overpressure due to the jet impact must be of the order of the time taken by a pressure wave to traverse the jet in the transverse direction. If, on the basis of published computational results, the jet radius r is estimated to be about 10% of the minimum bubble radius, ~ 1 μm or less, and the pressure wave speed $c \sim 2{,}000$ m/s, we have $r/c \sim 50$ ps or less.

Just as it is difficult to nucleate a bubble in a pure liquid, so it is to initiate a crack in a perfect crystal. In the usual fracture processes, cracks initiate at defects or other ''weak spots'' in the solid. As a matter of fact, fracto-luminescence in some solids has been reported to increase many times after exposure to x-irradiation that produces a large number of defects.[27] The observed strong sensitivity of sonoluminescence to small amounts of dissolved noble gases[7] may perhaps be explained on similar grounds. Since all the multipole moments of a noble gas atom vanish, at most only very weak bonds with the surrounding water molecules are possible. An argon atom could therefore function as a weak spot favorable for crack initiation. The xenon atom is larger than the argon one, the spot weaker, and crack formation easier. Conversely, helium is smaller and the tendency to crack formation inhibited. Nitrogen and oxygen molecules have a nonzero quadrupole moment, the binding with water is therefore

stronger, the weak spot not as weak, and cracking not as likely. The implications of these considerations for the intensity of luminescence are borne out by experiment.[7] If too many weak spots are present (high noble gas concentration), the material becomes friable and the process of crack opening may not be as energetic and disruptive of the liquid structure. This consideration might explain the luminescence intensity reduction in going from 1% to 100% argon concentration.[7] Another factor to keep in mind in this connection is of course that the bubble dynamics is strongly affected by the large difference in the value of the specific heat ratio between monatomic and diatomic gases.

It may also be relevant here to recall the existence of *clathrates*, compounds formed by the inclusion of molecules in cells that are present in some crystal lattices due to the particular geometry of the molecular structure.[32–34] Interestingly, although other types exist, the ability to form clathrates is a peculiarly characteristic property of water, and the xenon hydrate is one of the easiest-to-prepare and most widely investigated clathrates.[32] It will be recalled that xenon is also the ''best'' gas for sonoluminescence.[7] Although clathrates occur in the solid phase, it would seem quite reasonable that clathrate-like structures continually form and disintegrate in liquid water due to molecular agitation. Furthermore, some clathrates are stabilized by pressure, and, up to a point, high pressures promote the formation of clathrates even in some systems that show no clathrate formation at normal pressure.[34]

In conclusion I add a few considerations that reasons of space prevent me from developing more fully:

(1) Cooling the liquid has the effect of increasing the number of hydrogen bonds, or increasing the relaxation time for flow. Cracking is therefore more likely, and light intensity should increase in agreement with observation.[5] Since deuterium confers to heavy water more structural order than normal water (see, e.g., Ref. 35), one would expect differences between the two liquids. Unfortunately, current data are inconclusive, although they show the dramatic effects of minute impurities that would affect the liquid far more than the vapor.[36]

(2) Since 1-butanol is known to be surface active,[37] and therefore to adsorb at the bubble surface, the initiation of sonoluminescence at the surface, rather than in the bulk, might explain why the addition of as little as 40 ppm of this alcohol in water reduces the light intensity by a factor of nearly 25.[10] It may also explain the observed increase in jitter and rise time associated with aging of the water.[4]

(3) Field and co-workers have published high-speed photographs of the shock-wave-induced collapse of cylindrical cavities.[38–40] Some of these photos show an intense luminescence around the point of jet impact and the surrounding gas areas, the latter of which persist over two consecutive frames, i.e., for times in excess of hundreds of nanoseconds. The most probable cause of this effect is compressional heating of the gas, a mechanism proposed a long time ago[41] to explain sonoluminescence effects and substantiated by theory.[42] The effect is evidently different from that responsible for single-bubble sonoluminescence, as pointed out by Crum.[2] In addition to compressional heating, jet impacts of the type hypothesized here are also likely to occur in multiple-bubble sonoluminescence, which might therefore be a combination of both mechanisms. Interestingly, the photos of Ref. 40 also show (unfocused) shock waves harmlessly rattling around the bubble.

(4) Luminescence can be observed without difficulty in shock-tube experiments when the shock intensity is sufficiently strong. Its duration is far too long to account for the observed brevity of single-bubble sonoluminescence.[43]

Several aspects of the mechanism hypothesized here can be checked experimentally. In the first place, since the jet is predicted to be directed vertically, one would expect a variation of the light intensity along ''meridians'' in a vertical plane. Preliminary results on this aspect of the phenomenon have recently been reported.[44] Second, since jet formation depends on translational velocity, which is increased by buoyancy, sonoluminescence intensity is expected to increase in an artificially increased gravitational field. Third, a considerable amount of information on the effects of various types of dissolved molecules on the structure of liquids is available.[45] For example, water/methane solutions are expected to be very different from carbon tetrachloride/methane solutions. Similarly, water/ammonia, water/carbon dioxide, and water/dimethyl sulfate should have very different properties. Experiments of the type reported in Ref. 39 in which high-velocity liquid jets are made to impinge on liquid surfaces could also be revealing under the proper experimental conditions. Finally, it is obvious that fracture and light emission should also occur in the hypervelocity impact of solid bodies on liquid surfaces, although direct observation of the phenomenon may be difficult.

If the present hypothesis is correct, the implications on the mechanisms underlying sonochemical effects can be quite far reaching and a host of new techniques (and possibly technologies) can be developed to exploit microjet effects advantageously.

ACKNOWLEDGMENTS

Many of the ideas described before were prompted by conversations with N. Bourne, J. Field, W. Proud, A. Walton, and S. Putterman. The author also gratefully acknowledges discussions with R. Hickling, K. Stebe, L. Hargrove, L. Crum, R. Roy, R. Apfel, M. Longuet-Higgins, J. R. Blake, M. Paulaitis, and G. Astarita. Special thanks are due to J. Zheng and H. Oğuz for their help in generating Figs. 1 and 2. This study has been supported by the Office of Naval Research.

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