The Timing of Sonoluminescence

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We measured the timing of sonoluminescence by observing laser light scattered from a single sonoluminescing bubble. We performed this measurement on 17.8 kHz, 13.28 kHz and 7920 Hz systems and found that the flash typically occurs 100 nanoseconds before the minimum radius, contrary to previous claims that the flash always occurs within a nanosecond of the minimum radius. These results contradict the hot models of sonoluminescence, which require the flash to occur within a nanosecond of the minimum radius. We propose a new model: that the flash results from the discharge of an excited cold condensate, formed during the adiabatic expansion of the bubble.

Introduction

Sonoluminescence is the transduction of sound energy into light. With current techniques, sonoluminescence is created by focusing powerful sound waves in water or other fluids. Small bubbles present in the fluid will glow if the sound field is strong enough and other conditions are appropriate, such as the amount and type of dissolved gas present, and the ambient temperature in the fluid. The glow appears continuous to the eye, but in fact is the result of a rapid flashing which occurs once per acoustic cycle.

Experimental Design and Procedure

Each of the three resonator flasks we constructed is a hollow quartz sphere with a narrow neck and thin walls, (see Figure 1). The 500 mL flask was fitted with a one inch cylindrical PZT ring epoxied to the bottom center, and was held on the test stand by the neck with a small lab clamp. The 1 liter flask was fitted with a two inch cylindrical PZT ring. Because of its larger weight, it was not suspended by a lab clamp, but by a rubber hose epoxied to the outside of the neck. The 5 liter flask was fitted with a three inch cylindrical PZT and suspended by a rubber hose as well. All three flasks also had small PZT disks epoxied to the side wall which functioned as microphones to monitor the response of the flask to the drive.

The experiment begins by preparing the water-glycerine-gas mixture. It is important to minimize the amount of dust in the fluid mixture, because dust particles interfere with the formation of the sonoluminescent bubble. So, in a clean room, we mix approximately 1.36 liters of glycerine and 6.63 liters of deionized water for a total fluid volume of 8 liters, in a large 9.5 liter mixing flask. We then add about one gram of potassium sorbate to inhibit the growth of mold in the fluid. We cap the flask with the rubber stopper, and then take it out of the clean room and bring it to the lab room where we do the rest of the experiment. (This lab room could be made completely dark, and was cooled to 15 Celcius.) We then degas the mixture for several hours with a vacuum pump while stirring. After the fluid has been degassed, we add about 1/5th atm of argon gas above the fluid, and allow it to stir vigorously for at least 24 hours. After this, the fluid has been infused with argon and is ready to be used.

Next the fluid is poured into the resonator. To do this, the top of the mixing flask must be open to the atmosphere to allow the fluid to flow thru the stopcock and narrow tube into the flask. After the pouring is complete, the vacuum pump is used to remove the air above the fluid, the low pressure argon atmosphere is restored, and the remaining fluid is stored for later use.

The resonator flask is then placed on the test stand, and the audio power leads and microphone leads are connected. The first step is to find the exact resonant frequency of the flask so that bubbles can be levitated in the center of the flask. From a priori calculations, we know approximately what this frequency should be, so we set the generator to this frequency, set the drive of the generator to about 1.5 Vpp, and then turn on the power amplifier to about one third of max. Next, we adjust the value of the inductance to optimize the impedance match between the amplifier and the resonant circuit. Then we turn the power amplifier up to about two-thirds of max, and attempt to trap a bubble by disturbing the surface of the water with a syringe. We continue to adjust the drive and frequency until a bubble can be made to levitate in the exact center of the flask and cavitate. (When a bubble begins to cavitate it gets a 'fuzzy' appearance). At this point, we turn the room lights off and make further minute adjustments to the frequency and drive until the bubble begins to visibly glow. The 500 mL resonator has a resonant frequency close to 17.8 kHz, the 1 L resonator has a resonant frequency close to 13.28 kHz, and the 5 L resonator has a resonant frequency close to 7920 Hz.

To study the oscillation of the bubble and the timing of the SL flash in the bubble cycle, we scattered laser light from the bubble and monitored the scattered light signal with a lens, photomultiplier tube and oscilloscope (see...
This experiment is similar to previous micr-
scattering experiments. \cite{3}, \cite{7}, \cite{8}. The laser used was a
0.95 mW red HeNe. A large 4.5 inch diameter objective
lens with a 10 cm focal length was used to focus scattered
light from the bubble onto a Hamamatsu R647-25 Select
PMT with a 2.5 ns rise time. The PMT was run at
-850 Volts. The microphone PZT, sync signal from the
generator, and the output of the PMT were all monitored
and recorded with a Tektronix TDS 5034B oscilloscope
which has a sampling rate as high as 3 GHz.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1.png}
\caption{The design of the 500 mL, 1 L and 5 L resonators.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure2.png}
\caption{A schematic of the laser scattering experiment.}
\end{figure}

**Results**

Based upon repeated observations we find that the
sonoluminescence flash typically occurs 100 nanoseconds
before the time of minimum bubble radius. The sequence
of events is as follows: the bubble at first maintains a
steady radius for about 50% of the period. Then it sud-
denly expands to a larger radius, reaches an apex, then
begins to collapse. While collapsing, it emits a brief flash
of light, then that light is extinguished, whereafter the
bubble continues to shrink further, for an additional 100
nanoseconds, and sometimes longer. Finally, the bubble
reaches a distinct minimum radius, rebounds, bounces
several times, and then stabilizes. The cycle repeats from
there.

See Figures 4 thru 11 for examples. We have left these
oscilloscope traces as pure data, unaugmented with any
markings except the scale of the voltage and time axis.
We encourage the reader to take a straight edge, obtain
the scale, and verify our claims. Note that the photo-
multiplier tube that we used outputs a negative voltage
which is proportional to the cross-sectional area of the
bubble, so that these graphs are inverted.

**17.8 kHz Data**

Figure 4 depicts one cycle of 17.28 kHz sonolumines-
cence. It is an average of eight traces. In this figure, sev-
eral bounces are clearly visible, of at least three distinct
minimum radii, proving that the motion of the bubble
is stable over several periods and that timing informa-
tion is not lost due to oscilloscope averaging. Thus when
we look at Figure 5, which is a zoom-in near the first
bounce, we see that the sonoluminescent flash, visible as
the upside down spike, precedes the minimum radius by
over 100 nanoseconds. Since this spike is only slightly
broader than the 10 nanosecond width of a single-shot
trace of a flash on our system, \cite{8}, we know that we see
compelling evidence of several flashes that repeatedly and
regularly occurred over 100 nanoseconds before the min-
imum radius.

Figure 6 shows two full bubble periods. The zoom-
in in Figure 7 is on the first flash, which is somewhat
broader due to averaging, but still distinct from and
preceding the minimum radius by at least 100 nanosec-
onds.

**13.28 kHz Data**

Figure 8 shows the advantage of viewing the data on
a large scale before zooming in near the minimum ra-
dus. Notice how well defined the first minimum radius
bounce is. Three or four data points appear to point
to it distinctly. We should keep this in mind when we
look at the zoom-in shown in Figure 9, which at first ap-
ppears less definitive, unless we identify the three points
of the minimum radius from the zoom out, measure the
difference between the center of these three points and
the beginning of a broad flash which exceeds the scale
of the zoom. Again, a time difference in excess of 100
nanoseconds is apparent.
Hypost of Condensate Energy

These experimental results imply that the compres- shock heating model of Greenspan, Wu, or others like it that require the flash to occur within one nanosecond of the time of minimum bubble radius must be ruled out. We present a model which is consistent with the fact that the flash occurs 100 nanoseconds before the minimum bubble radius, at times when the bubble wall motion is subsonic, and temperatures and pressures are near standard conditions.

Now, during the acoustic cycle an oscillating bubble is both expanding and contracting. The calibration of the laser scattering data, as reported in previous experiments [1], [2], [3], [8], [7], implies that the maximum radius of the bubble is larger by a factor of ten compared to the equilibrium radius. This is a volume expansion ratio of 1000. Consider that during adiabatic expansion, the temperature of a van der Waals gas drops as \( \frac{1}{V(\gamma-1)} \). A radial expansion factor of ten will result in a hundred-fold decrease in temperature, \( \gamma \) being 5/3 for a monatomic gas such as argon. Thus if the gas is at room temperature at the equilibrium radius, it will be cooled to a temperature below 4 Kelvin by the time it reaches the maximum radius – cold enough to liquify or freeze any gases in the bubble. When the bubble subsequently collapses, the contents of the bubble will warm back up, and any condensate formed during the expansion will be destroyed by the time the bubble returns to its equilibrium radius, which will occur approximately 100 ns before the time of minimum bubble radius, (see Figure 2).

During adiabatic expansion, the gas in the bubble will cool as a result of a process working on its surroundings. The pressure of the gas will drop as
\[
P(V) = P_0 \frac{(V_0 - nb)\gamma}{(V - nb)\gamma}
\]
Now we can calculate the work done by the gas during an adiabatic expansion. We expect the integration of \( P(V) \) to infinity to be convergent, and that the amount of work that can be performed by a gas on its surroundings, the \textit{adiabatic work capacity} \( W_{\text{adiabatic}} \), is finite.
\[
W_{\text{adiabatic}} = \int_{V_0}^{\infty} P(V) \, dV = \frac{c_v}{R} P_0 (V_0 - nb)
\]
Here, \( R \) is the ideal gas constant, \( c_v \) is the molar constant volume heat capacity, \( n \) is the number of moles, and \( b \) is the van der Waals excluded volume per mole. The term in \( P_0 V_0 \) is the work that could be performed by an ideal gas, which is reduced by the van der Waals term \( \frac{c_v}{R} P_0 nb \), which is the energy reserved for condensation.

Let us discuss this concept of condensation energy further. When a gas has been cooled to a temperature infinitesimally close to its condensation point, just before it condenses, there is energy present in the gas because the gas atoms are “bouncing around” with center of mass motion. Once it spontaneously condenses, it forms droplets which are relatively stationary. So then where did that energy go that was in the center of mass motion? Well, it must be conserved, and to do so, the energy is transferred into excited electrons in the condensate. The van der Waals empirical model implies that the term \( \frac{c_v}{R} P_0 nb \) represents the energy stored in a van der Waals condensate. For a bubble of equilibrium radius \( R_0 \) and temperature \( T_0 \) we can use the equation of state to replace the number of moles of gas \( n \) in the condensate energy expression:
\[
E_{\text{condensate}} = \frac{4 \pi c_v P_0^2 R_0^3}{3 R RT_0} b
\]
Eq. (3) is a useful formula to the experimentalist. It predicts that a 10 \( \mu \)m bubble starting at room temperature will form a condensate which stores approximately 1 picoJoule of energy. Calibrated measurements of SL flashes from bubbles this size, [9], have shown that each SL flash contains about 1 picoJoule of energy. Eq. (3) also predicts that cooler ambient temperatures will produce brighter bubbles, that larger bubbles will also be brighter, and that higher static pressure will produce brighter bubbles. These predictions have been observed.

For a 10 \( \mu \)m bubble such as is typical, this formula predicts an energy of 1 picoJoule per flash, which is equivalent to 7 MeV. From a macroscopic scale, 1 picoJoule doesn’t seem like a lot of energy, but 7 MeV is a lot of energy.
on the electron scale, especially if that 7 MeV is stored in one or a few electrons – in which case the discharge would certainly produce the bremsstrahlung spectra seen [10].

Note that this formula predicts that larger bubbles will have more energetic discharges. If one were to trap 100\(\mu\)m bubbles in larger, lower frequency resonators, condensate energy could exceed 7 GeV. This might be enough energy to provoke a fusion reaction in the bubble if the right isotopes are present, in addition to a much more visibly powerful discharge.

**Conclusion**

Based upon our understanding of thermodynamics, we believe that the rapid expansion of the bubble suddenly cools the interior atmosphere to a few degrees Kelvin – cool enough to liquefy even helium or hydrogen. This sonically induced condensate must store the latent heat of the previous vapor in internal electronic excitations. The condensate is meta-stable and persists until the collapsing bubble wall gets close enough for an electric arc discharge of several MeV to occur between the condensate and the bubble wall.

We agree with the statement that sonoluminescence is an energy focusing phenomenon. But our understanding is that the energy of the discharge is primed by the rapid cooling of the gas, rather than a compression shock-plasma as suggested by others. For that mechanism requires high bubble wall velocities which only occur within one nanosecond of the minimum radius. This is ruled out because we observe flashes at times 100 nanoseconds prior, when the bubble wall is still moving slowly.

One last comment about the absence of any statistical analysis of the flash timing in this work. The thesis of this article is a refutation of the claim by others that the sonoluminescence flash *always* occurs within a nanosecond of the minimum radius. To disprove their claim requires only a *single* counterexample, while we have presented several. A statistical analysis of the mean and standard deviation would teach us more about how the timing varies, but it is not necessary just for refuting the claim that the flash time is always precisely synchronous with the time of minimum bubble radius.

[1] S.J. Putterman and K.R. Weninger. “Sonoluminescence: How Bubbles Turn Sound into Light.” *Annual Reviews of Fluid Mechanics* 32 (2000): 445-476.
[2] M. Brenner, S. Hilgenfeldt and D. Lohse. “Single-bubble Sonoluminescence.” *Reviews of Modern Physics* 74 (2004).
[3] B.P. Barber and S.J. Puttermann. “Light scattering measurements of the repetitive supersonic implosion of a sonoluminescing bubble.” *Physical Review Letters* 69 (1992): 3839-3842.
[4] K.R. Weninger, P.G. Evans and S.J. Puttermann. “Time correlated single photon Mie scattering from a sonoluminescing bubble.” *Physical Review A* 61 (2000): 1020-1023.
[5] Harvey P. Greenspan and Ali Nadim. “On sonoluminescence of an oscillating gas bubble.” *Physics of Fluids A* 5 (1993).
[6] C. C. Wu and Paul H. Roberts. “Shock-Wave Propagation in a Sonoluminescing Gas Bubble.” *Physical Review Letters* 70 (1993).
[7] T.E. Brennan. “On the Variation of Sonoluminescence Flash Timing.” Ph.D. thesis. Illinois Institute of Technology.
[8] T.E. Brennan. “Variations in Sonoluminescence Flash Timing.” http://arxiv.org/abs/1012.5009
[9] D. Felipe Gaitan, Anthony A. Atchley et al. “Spectra of single-bubble sonoluminescence in water and glycerin-water mixtures.” *Physical Review E* 54 (1996).
[10] Robert Hiller. Ph.D. Thesis. UCLA 1995.
FIG. 4: One cycle of a 17.8 kHz argon SL.
FIG. 5: Zoom-in on previous.
FIG. 6: Two cycles of a 17.8 kHz argon SL.

17.8 kHz Argon SL: /17.8kHzSLData/110707_001028.csv
FIG. 7: Zoom-in on first flash in previous.
FIG. 8: One cycle of 13.28 kHz argon SL.
FIG. 9: Zoom-in on previous.

13.28 kHz Argon SL: /13.28kHzData/110719_000003.csv

PMT Voltage

ns Time

-0.0005

-0.0010

-0.0015
FIG. 10: One cycle of 7920 Hz argon SL.
