SONOLUMINESCENCE: SOUND INTO LIGHT

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Abstract

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Single Bubble Sonoluminescence or SBSL is the phenomenon in which light is produced from an acoustically driven collapsing bubble trapped in a fluid. This is possible to achieve in a laboratory setting with relatively inexpensive equipment and this experiment is the basis for this paper. In this paper I will discuss the method used to achieve SBSL, various other similar experimental results and the competing theories used to explain the method by which the light is produced.

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I dedicate this thesis to my Mother.

Who is a Superhero.

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1. Introduction:

Sonoluminescence, literally: light from sound, is the process by which acoustic waves cause a nonlinear, rapid collapse of a gaseous bubbles in a liquid medium which results in a very brief flash of light. The light flash is on the order of picoseconds and this process results in a concentration of the ambient acoustic energy by 11 orders of magnitude. This is a highly nonlinear situation resulting in an extreme phenomenon, which isn't fully explained and is an interesting and active area of research.

The first documented case of sonoluminescence occurred in 1934 at the University of Cologne. Two researchers, Frenzel and Schultes discovered it by accident while trying to speed up the photographic developing process by putting an ultrasound transducer into the photographing developing fluid. They noticed tiny dots on the film and concluded that sonoluminescence was responsible. However the discovery is also accredited to Marinesco and Trillat in 1933 "when photographic plates submerged in an insonified liquid became exposed. This was then left unexplained until 1947 when Paounoff realized that this exposure was occurring at the pressure maxima in the liquid." [1]

Up until 1987 most all of the sonoluminescence known was later to be categorized as Multi Bubble Sonoluminescence or MBSL. Clouds of erratically behaving bubbles characterize this phenomenon. Therefore the theory to explain MBSL can only be based on averages and blunt estimations. Theories used to explain sonoluminescence were lacking until 1987 when Felipe Gaitan from the University of Mississippi discovered the necessary parameters for achieving stable SBSL. Gaitan was able to trap a single bubble in a glass cylinder and was able to produce periodic and stable bubble collapses and resulting light flashes by applying sinusoidally varying acoustic pressure waves via piezoelectric transducers attached to the ends of a cylinder. The light flashes were emitted once per acoustic cycle, lasting around 100 picoseconds and consisted of blue/white light. The cycles repeated with nearly perfect periodicity. Soon, however, Gaitan refocused his efforts on what he considered more important work. The torch was then passed to Bradley Barber, Bob Hiller and Seth Putterman, who during the mid nineties did most of the important quantitative measurements on the mechanics of the bubble, the gas content in the liquid and the analysis of the spectrum of the light emitted. With these experimental measurements, many other scientists have become involved to explain the theory behind the phenomenon that can fully explain all of the data collected. To this day no one theory is universally accepted and exciting research is ahead in this respect.

Sonoluminescence belongs to a larger phenomenon called Noninertial Cavitation, which is defined as "the process in which a bubble in a fluid is forced to oscillate in size or shape due to some form of energy input, such as an acoustic field." [2] Lord Rayleigh was the one who originally studied cavitation, wherein he formed a mathematical theory to model the collapse of a spherical bubble of gas in a liquid medium. Milton Plesset later revised Rayleigh's theory to form what is now known as the Rayleigh-Plesset (RP) equation for the radius of a bubble over time (R(t)). This comes from the Navier-Stokes equations, which are a set of nonlinear differential equations that model the flow of liquids. More recently, various theorists have applied modifications to the RP equation to describe the collapse of a bubble as seen in SBSL however the theories fail during the final stages of the collapse and thus the reasons for the actual light emitting process are still heavily debated.

The particulars of the theory are beyond the scope of this paper and thus this paper focuses mostly on the actual experiment with some acknowledgment of the various theories that exist to explain it. More relevant to an undergraduate student is the experimental methods used to achieve SBSL and the various other extensions to the basic experiment that have been done in the past two decades.

2. The Experiment:

This experiment is based on an article in Scientific American by Hiller and Barber entitled *The Amateur Scientist, Producing Light From A Bubble Of Air*, [3] with some advice taken from a paper written by W.A. Steer entitled *Sonoluminescence experiment: Sound Into Light*. Steer recreated the exact same experiment as Hiller and Barber but went into much more detail after he found the Scientific American article to be much too vague in more than a few respects. This is the roughly same set-up used by Felipe Gaitan in 1987 and has remained fairly similar throughout all of the SBSL experiments done since then. Of course, in more sophisticated labs, they also use various spectrometers, LASERs and Photomultiplier Tubes (PMT's) to obtain direct measurements of the radius of the bubble as a function of time (R(t)) and also of the spectrum and intensity of the light emitted. The apparatus used, shown below, consists of only the equipment necessary to view the bubble collapse and resulting light emission, no actual measurements of the phenomenon were taken due to time constraints and the fact that the equipment was unavailable to an undergraduate student at UCSC.

2.1 Apparatus



Figure 1. The experimental apparatus used.

What you have above is a resonant (LRC) circuit wired to an acoustic resonator. The goal is to

match the acoustic resonance with the circuit's electrical resonance to achieve maximum power to the

bubble in the flask. A list of equipment used in the experiment appears below as well as a circuit

diagram illustrating the resonant circuit.

Equipment used:

- 1. 100ml spherical boiling flask.
- 2. Two 20mm x 6mm drive piezoelectric transducers.
- 3. 1 3mm x 1mm microphone piezoelectric transducer.
- 4. 1 ring stand with attached three-finger clamp.
- 5. 34 gauge and 36 gauge wire.
- 6. Coaxial cables.
- 7. Banana Clips.

- 8. An oscillator.
- 9. A Tektronix oscilloscope.
- 10. An audio amplifier.
- 11. A 25mH inductor.
- 12. A 500ml spherical boiling flask.
- 13. A hot plate for boiling the water.
- 14. Rubber stoppers and tubing to create a vacuum over the degassed water.
- 15. 5-minute epoxy.
- 16. 1 Ohm, 10 KiloOhm and 1 MegaOhm resistors.



Figure 2. The circuit diagram representing the experimental apparatus.

2.2 Experimental Methods:

2.2.1. Set-up.

It was necessary to order the 100 and 500ml flasks as well as the piezoelectric transducers, which were available as a package from a source cited in the article. After two weeks of waiting for the supplies, the experimenters were ready to start and the first step in assembling the apparatus was to solder the lead wires to the drive and microphone transducers. The transducers are polarized and therefore they must be attached to the flask in the same orientation so that they are synchronized in their rarefactions and contractions. Piezoelectric transducers are designed so that when current is applied to them, they respond by physically expanding or contracting based on the sign of the derivative of the applied current. Therefore, when a sinusoidal current is applied they will expand and contract sinusoidally which creates the necessary standing pressure wave in the flask for sonoluminescence. Three leads were soldered to the positive side of all three transducers and these sides were glued to the flask to minimize stress and avoid confusion. The drive transducers were glued to the equator of the spherical flask, directly opposite each other. The microphone transducer was glued directly on the south pole of the spherical flask, and this was used to monitor the acoustic waves in the flask. Applying current to the transducers causes expansion but the opposite is also true, which is why the resonating flask causes the microphone to send current to the oscilloscope.

Once the wires were connected securely to the transducers and they were epoxied to the flask the electronics were then set up as in Fig. 2.1. A standard desk was used as a platform for all of the equipment and the wired connections from the transducers were shielded in heat shrinking plastic and electrically taped to the ring stand to prevent any unnecessary stress on the connections. The output of the microphone was connected to channel 1 of the oscilloscope and was the main source of feedback to the experimenters. Once the set-up was complete, the next step was to establish acoustical and electrical resonance. 2.2.2 Proper Fluid & Resonance.

The acoustic resonance necessary for SBSL creates a standing wave in the flask with the pressure maxima, or anti-node, at the center of the flask. The wavelength of this pressure wave will therefore be the diameter of the glass and the frequency will be approximated by Eq. 2.2.

Equation 2.2: f = v(T)/d

where f is the resonant frequency, v(T) is the velocity of the sound in water as a function of temperature and d is the diameter of the flask.

The electrical resonance was estimated by Eq. (2.1), the approximation of the resonance of an LRC circuit.

Equation 2.1:
$$\omega = 1/(LC)^{0.5}$$

With a frequency calculated to be near 25 kHz, the inductance therefore needed to be around 20 mH and this was a good starting point for achieving SBSL. The frequency is raised from the calculated value in Eq. 2.2 by ten percent to account for the change in velocity of the sound waves through the glass.

To achieve stable SBSL, one must use sufficiently de-gassed water so that there will be less extraneous forces on the bubble in the center of the flask. The water is de-gassed by boiling it and then immediately stopping the flask with a rubber stopper. The induced constant volume will create a vacuum over the water as the temperature cools and the vapor condenses. This water is then poured along the wall of the flask to introduce a small amount of air, about twenty percent of the normal standard concentration. Once the flask is full one must view the output of the current and voltage and adjust the inductor until they are in phase, this ensures that the circuit is in resonance, electrically. The frequency is then adjusted until resonance of the flask is observed by viewing the microphone output on the oscilloscope. The resonance for our experiment occurred around 28.5 ± 0.2 kHz and then the inductor is adjusted further to amplify the signal as seen from the microphone.

2.2.3 Achieving SBSL.

The system was tuned to resonance and then, using a dropper, a small amount of water was extracted and splashed onto the surface of the liquid to introduce bubbles in the flask. When the system was tuned extremely well bubbles were then created by simply dabbing the top of the water in the flask with a glass rod. This would introduce on average three or four bubbles that would coalesce into the center and react differently based on the amplitude and frequency of the driving acoustic waves. The experimenters were able to trap the bubbles in the center and obtain perfect stability, which was an obvious sign of resonance. Then, as instructed in the *Amateur Scientist* article they would slowly increase the amplitude to achieve the characteristic periodic compression and rarefaction. Increasing the amplitude would cause the bubble to react exactly as described in the Hiller/Barber and Steer articles. The bubble would start to jitter over a few millimeters, then become stable again. Then it would shrink as described and at 28.47kHz and with an amplitude of 385mV light was produced. It was a bright bluish white light that repeated so fast it seemed almost like a continuous flash. It lasted over 30 seconds and then the experimenter began to increase the output of the amplifier and extinguished the light.

The experimenters tried to create SL by varying the boiling time and cooling time of the water, and did notice that with colder, more de-gassed water, the bubble was a lot more stable. There was

difficulty in using the colder water because the resonant frequency would change as the water "aged" because the speed of sound in water is dependent on the temperature. These changes were taken into consideration and many frequencies around the resonance (varying up to 2 kHz) were tried. The experimenters noticed that the water used in this experiment needed to be boiled for much longer than the expected time, which might be due to the fact that the hot plate took a long time to create a fully rumbling boil of the water. Also the water seemed to age very fast, SL was only observed one time and due to time constraints was not replicated because of the time it takes to prepare the water correctly. The experimenters struggled for weeks to create SL getting all but the light to work and the crucial variable holding them back was the gas content in the water. After tripling the boiling time used in the previous attempts, SL was finally achieved.

2.2.4 Possible reasons for error.

At first the experimenters made an inductor by wrapping wire around a plastic spool a few thousand times and this was gradually reduced until maximum amplitude was achieved. However changing the amount of wire seemed to have a nonlinear and irregular effect on the amplitude of the microphone output so a static 25 mH inductor was substituted and electrical resonance became immediately apparent around the 27 - 29 kHz range. However the lack of variability in the inductor could have been a possible source for the lack of precision and the difficulty to achieve light.

In both the articles this experiment was based on the lead wires that were soldered onto the transducers were sheathed and connected to coaxial cables within a few millimeters. In the set up, shown in the picture below, the reader can see that the 34 gauge lead wires were tens of centimeters long and were soldered to much thicker copper wire. These connections were protected with heat shrinking plastic and were taped to the ring stand to minimize the stress on the connections. However

there was apparent cross talk between the exposed lead wires, especially at high amplitudes. Often the system would short circuit if the amplifier was turned to higher power levels and the signals would cut out to much annoyance of the experimenters.

The sphericity of the flask was also a problem duly noted in the Steer article. He had to go through more than one flask to achieve SL so that could have been holding back the light emission; however with the amount of power the experimenters had available it is does not seem like that big of a source of error.

Another concern to the experimenters was the oscillator. Three were used in total and the first two were not sufficient due to their precision of only a few kHz and their obvious instability. The third oscillator was actually a pulse generator but seemed to produce almost an indistinguishably sinusoidal pattern of pulses and was much more precise and stable (to 1 Hz/mV). However the fact that it was not actually a function generator could have also caused the bubble to react adversely. The knobs on all the oscillators would sometimes react strangely, jumping at almost random intervals and reversing the +/- spin directions. This was frustrating and made it almost impossible to stably increase the frequency and amplitude.

"The gas content is a determining factor of whether or not the bubble will survive at the required acoustic amplitude. Qualitatively, with increases in acoustic amplitude (and even without), overly degassed water will absorb the bubble. The bubble shrinks to death." [4] This was by far the thing holding the experimenters back the most.



Figure 3. A snapshot of the experimental apparatus.

3. Relevant Experimental Results.

Various factors must be taken into account when studying sonoluminescence. Research from the past two decades has shown that SBSL is only viable within a small range of parameters and can greatly be enhanced with changes to certain highly sensitive variables. Light is only emitted if everything is tuned to within very precise ranges, but the intensity and spectrum of the light can greatly vary by changing certain inputs. This is a great way to gain insight into the mechanisms behind SBSL and is an active area of research. Also, using lasers and Photomultipliers to track the radius of the bubble as a function of time has been a big problem during the end stages of the collapse, which is where classical bubble theory fails. In this section the various experimental advances will be discussed.

3.1 Radius During Collapse.

Once Gaitan had successfully achieved stable SBSL he began to measure the bubble's collapse using a technique called MIE scattering. His followers then used the same technique with more precise equipment to obtain better data. This is explained in the quote below:

"CW light scattering methods have been effective in measuring the dynamical response of the bubble to the driving sound for all phases of the motion except near the light emission. The supersonic collapse requires sub ns time resolution over a large dynamic range of signals, transcending the capabilities of the typical photomultiplier tubes (PMT) used to detect the scattered light. Previous work used fs pulsed laser scattering to overcome the 2 ns response function of detection PMTs to measure this bubble motion, but was limited by the 2 GHz sampling speed of the digitizer recording the signal. Here we report on a new technique combining pulsed Mie light scattering with time correlated single photon counting (TCSPC) for detection of the scattered light which here yields about 50 ps time resolution of the supersonic bubble collapse." [5]



Fig. 9. The radius R(t) scaled to the maximum radius R_m during one cycle of the sound field. If V(t) is the signal on the photodetector and $\tilde{V}(t)$ is the noise level, this plot is given by $\{|[V(t) - \tilde{V}(t)]|/V_m\}^{1/2}$, where V_m is the maximum voltage. Note that for a SL bubble $R_m/R_0 \approx 10$, where R_0 is the ambient radius which is the radius the bubble would have in the absence of a driving pressure. The driving sound field is superimposed. This figure is constructed from the data in Fig. 8, for an air bubble in water. A comparison to the Rayleigh-Plesset equation of bubble dynamics indicates that for these data $R_0 = 4.0 \ \mu$ m, and the amplitude of the sound field is 1.35 atm.

Figure 4. Radius versus time during one cycle of the bubble collapse

Mie scattering is defined as: "is an analytical solution of Maxwell's Equations for the scattering of electromagnetic radiation by spherical particles." [6] Therefore by using the techniques describe above, Gaitan and then Peterman/Barber/Hiller were able to plot the radius versus time of the bubble decay as shown in Fig. 4 and Fig. 5. The pattern in Fig. 4 is repeated almost perfectly periodically, with measured variances of less than 50 picoseconds. The bubbles' radii oscillate between 4 and 40 microns in similar experiments, however this can vary with gas type, content, and the partial pressure above the liquid medium. The consistency with theory of this aspect of research will be discussed in section 4.



Figure 5: Radius versus time during the collapse of a bubble undergoing SL.

From Fig. 5: "we can conclude that the bubble is collapsing inward at Mach 4, relative to the ambient speed of sound in the gas, and that the acceleration which brings the bubble to a halt at its minimum radius exceeds 10g (Barber et al., 1997). This acceleration implies that over 90 percent of the acoustical energy inputted into the bubble is expelled as a single shockwave, which further limits theory in this region. In addition, as shown in Fig. 13, the SL light flash is localized to within 250 ps of the minimum."

These data show the astounding focusing of energy that occurs in SBSL and the next logical step is the analysis of the light produced.

3.2 Light Spectrum.

The flashes of light produced from SBSL can last anywhere from 35-380 picoseconds. These flashes emit photons that contain energies above 6 electron volts, which correspond to temperatures in the tens of thousands of degrees Kelvin (70,000 K). This has led to the leading (plasma) theory explaining the light emission and also to the possibility of creating fusion through energy focusing of this kind. The most amazing part is that the length and width of the flashes are constant regardless of

the wavelength. The spectrum emitted is broadband with wavelengths ranging from the infrared to the ultraviolet, limited by the cutoff wavelength imposed by water of around 200nm. The spectrum of light produced above waters' cutoff frequency is one of the important unexplained characteristics of SL.



Figure 6. The flash widths produced by SL and also the time each wavelength of light is emitted. The longer emission of light below 320 nanometers is attributed to the differences in the indexes of refraction and also the vessel used. [8]

Another interesting addition to the study of SL is the type and concentration of gas dissolved in the water. Once early experimenters realized that air was the best medium for SL to occur in they began to try and create more intense light output using purer media. However, they found out that by partially recreating air using an approximation of 80% Nitrogen and 20% Carbon, no light was emitted. It became apparent that the crucial difference between these two gas mixtures was that fact that air has approximately one percent Argon in it and then it these researchers realized that doping the medium with a noble gas was necessary to produce light. Pure noble gas mixtures were then tried with no success so therefore it is necessary to have a very low percentage of a noble gas (1-2%) for light to even be possible. The rest of the gas can be a variety of things but that small percentage of noble gas is crucial. Shown below is the variance in the spectral radiance of the light emitted based on various gas compositions.



Figure 7. Spectral radiance vs. wavelength of light emitted based on dissolved gas composition.

One of the tougher areas of research in SBSL is the study of the sphericity of the bubble near its

minimum radius. In a paper by Weninger/Barber/Putterman entitled *Angular correlations in sonoluminescence: Diagnostics for the sphericity of a collapsing bubble*, the authors use multiple PMT's to describe the angular correlations in SL. Basically they discover a dipole component in the detected photons and state that:

"The long time decay of the angle dependent correlation indicates that the dipole component is due to some aspect of the hydrodynamic motion. Such motions rearrange themselves on the same time scale for which the sound field changes. Various possibilities include i. Jitter in the location of the bubble, ii. bending of the emitted light by the sound field in the bulk of the fluid, and iii. Refraction of the SL rays by the surface of a nonspherical bubble." [9]

However the authors eliminate i. and ii. as possibilities because of the uniformity of the light detected and go on to state that the characteristic angular correlation measured in the experiment is possibly due to the nonsphericity of the collapse of the bubble. They also show how their data are consistent with an elliptical refraction of the light and go on to describe the various degrees of correlation between the dipole component and the percentage of ellipticity.

"According to our interpretation the observed dipole provides a probe of the degree of nonsphericity of the collapse. Ellipticity is the leading order, quadrupolar, form of a convolution instability. Such instabilities have been studied with regard to bubble and shock wave motion and inertial confinement fusion. They have also been implicated in the upper threshold of SL. Another type of asphericity that occurs in a collapsing bubble is the formation of a jet."

Thus this research sets the stage for a more complete and accurate theory of the bubble collapse and light emission found in SL.

4. Theory.

One cannot jump into current theories of SL without first describing bubble dynamics classically, which is precisely what Lord Rayleigh started in 1917. Rayleigh was motivated by cavitation damage observed in Naval propellers and created a theory to describe the isothermal collapse of a spherical

bubble trapped in a liquid medium. Most of the current theories are extensions of the Rayleigh-Plesset equation and become convoluted very fast. Plesset applied Rayleigh's theory to a bubble in an acoustic sound field and within certain parameters, which 99.5% of this experiment falls in; it can accurately describe the bubble dynamics.

The parameters necessary for this equation to be accurate are listed as follows:

In the limit where the imposed sound field $P_a(r, t)$ has a small Mach number, such that $|P_a/\rho c^2| \ll 1$, where c is the speed of sound in the fluid (c_g will denote the speed of sound in the gas), and the Mach numbers and accelerations of the bubble are small,

$$rac{\dot{R}}{c}\ll 1, \quad rac{R\ddot{R}}{c^2}\ll 1, \quad rac{\dot{R}}{c_g}\ll 1, \quad rac{R\ddot{R}}{c_g^2}\ll 1,$$

and the wavelength of the sound field, $\lambda = 2\pi/k$, is large compared to the bubble radius, $kR \ll 1$, one is led to the leading order Rayleigh-Plesset equation (Rayleigh, 1917; Plesset, 1949; Noltingk and Neppiras, 1950; Prosperetti, 1984; Prosperetti et al., 1988; Löfstedt et al., 1993)

$$R\ddot{R} + \frac{3}{2}\dot{R}^{2} = \frac{1}{\rho}(P_{g}(R) - P_{0} - P_{a}(0, t)) - \frac{4\eta\dot{R}}{\rho R} - \frac{2\sigma}{\rho R} + \frac{R}{\rho c}\frac{d}{dt}(P_{g} - P_{a}).$$
(5)

Figure 8. The RP equation and the necessary approximations to make it valid for SL

Modifications to this equation have led to a great fit for the data of the radius of the bubble during the acoustic cycle, showing only a six percent difference in the minimum radius (seen in Fig. 9), which is due to the neoclassical effects near the light emission region attributed to, for example, shockwaves, plasma and jets.



Figure 9. The comparison of the solution for the radius/time curve derived from the RP equation and experimental data.

If there was no light emitted the RP equation would perfectly describe the bubble dynamics, however in the final stages of the collapse there is a small but significant deviation from this theory and many scientists have been trying to account for this deviation.

The broadband spectrum produced from SL has puzzled scientists who originally tried to equate it to other "cold light" mechanisms such as frictional light emission and light emitted from fracturing crystals. The spectrum from these show sharp peaks in certain small wavelength ranges and is the reason for their characteristic colors. The lack of these sharp peaks leads scientists to believe that the light is produced not from "cold light" mechanisms but rather from very hot spots which has been confirmed experimentally by measuring the energy of the photons released.

The adiabatic process characterized by SL can in fact predict about 1/6th of the temperature reached within the bubble. Rayleigh assumed that the gas in the bubble remained at the same temperature throughout the process (isothermal) and therefore was unable to predict this phenomenon.

Some of the explanations for this discontinuity include blackbody radiation, *Brehmsstrahlung* and chemiluminescence.

A better theory for explaining the energy gap is the shock wave model. In this model a shock wave is formed at the final stages of the collapse, which increases the speed and amplitude of collapse. The shockwave has never been verified experimentally but remains one of the leading explanations for the extreme energy focusing. It leads to experimentally relevant results when an input collapse speed of Mach 4.3 is instituted (which is close to the measured Mach 4 collapse). It predicts a temperature of 10⁵ K, a minimum radius of 0.15 microns and a light flash of 100 picoseconds, all of which agree handsomely with experiments. The temperature this theory predicts in the bubble would lead to an ionization of the gas, creating plasma. The expelled electrons would then collide with the ions formed and would emit the light.

"The launch of a shock wave by the Rayleigh-Plesset bubble dynamics with the subsequent Bremsstrahlung radiation from the hot ionized contents is the most complete candidate model of SL. This model, however, is far from satisfactory. It requires a number of key physical inputs such as the acoustic drive level and ambient radius (Pa and *Ro*) at which bubbles give SL and additional physical inputs are required in order to determine the minimum radius attained by the shock. Furthermore, it does not address compelling yet simply stated mysteries, such as why pure diatomic gases are bad sources of SL."

Obviously theory has come a long way over the past 6 decades but it is far from complete and a complete explanation will be a monumental and exciting development in physics today.

5. Conclusions

The acquisition of light from a bubble trapped in a liquid medium is an amazing and still yet unexplained phenomenon. The energy focusing is extremely nonlinear but sonoluminescence is characterized by extremely accurate periodicity, where chaos would be expected. Research in the past two decades has unfolded a lot of evidence and theories supporting those. Lots of information however is still yet unknown and research in the future will be both groundbreaking and extremely exciting. The experimenters learned a great deal about bubble dynamics, circuits and acoustics from this thesis and were extremely impressed in obtaining light from sound. The author wishes future experimenters the best of luck in acquiescing the faint "star in the sky".

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